ISOTOPES AND RADIATION TECHNOLOGY

A Quarterly Technical Progress Review

Prepared for DIVISION OF TECHNICAL INFORMATION / DIVISION OF ISOTOPES DEVELOPMENT, UNITED STATES ATOMIC ENERGY COMMISSION, by P. S. BAKER, A. F. RUPP, and ASSOCIATES, OAK RIDGE NATIONAL LABORATORY

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Nuclear Safety, Wm. B. Cottrell, W. H. Jordan, and associates, Oak Ridge National Laboratory

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ISOTOPES AND RADIATION TECHNOLOGY

A REVIEW OF RECENT DEVELOPMENTS

Prepared for DIVISION OF TECHNICAL INFORMATION / DIVISION OF ISOTOPES DEVELOPMENT, UNITED STATES ATOMIC ENERGY COMMISSION, by P. S. BAKER, A. F. RUPP, and ASSOCIATES, OAK RIDGE NATIONAL LABORATORY

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Foreword

Isotopes and Radiation Technology is a Technical Progress Review prepared by the Isotopes Development Center, Oak Ridge National Laboratory, at the request of the Divisions of Technical Information and Isotopes Development, U. S. Atomic Energy Commission. This Review is intended to assist those interested in keeping abreast of significant developments in the fields of isotopes and radiation technology. It is not a comprehensive review of all literature published in this field during a given quarter; rather, it is a mechanism for presenting concise, selected reviews of information on subjects of prevailing Commission interest as it becomes available.

This Review attempts to relate the published results of research and development sponsored by the Division of Isotopes Development to significant developments in radioisotopes and radiation technology, as reported in the world literature. Coverage includes isotope production and development, isotope technology development, isotope applications technology, process radiation development, and radiation processing of food and medical supplies.

In addition to reviews of current literature and special review articles dealing with specific isotopes, facilities, and applications, this publication occasionally contains feature articles prepared by recognized experts on specific topics of current interest. Critical evaluations and interpretations presented are those of the editors and invited reviewers; therefore readers are encouraged to consult the original references in order to obtain all the background of the work reported and the interpretation of the results given by the original authors.

The subject headings listed below have been adopted to help maintain continuity and order, from one issue to the next, in the material reviewed. All reviews, except Feature Articles, will be arranged under these headings; but any one issue will not necessarily contain all the headings.

Isotope Production and Development
Isotope Technology Development
Food Irradiation
Process Radiation Development
Source Development
Isotopic Power Development
Applications in Medicine
Safety
Miscellaneous Items of Interest

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Section

I

Isotopes and Radiation Technology

General Review

One of the responsibilities of the editors of *Isotopes and Radiation Technology* is to provide worldwide coverage of those areas of isotope production and application which are of direct interest to the USAEC's Division of Isotopes Development (DID). This reporting is difficult since so many different organizations throughout the world are involved in the isotope business and their publications are issued irregularly.

Two possible approaches to the problem are (1) to cover one or more countries at a time and describe insofar as possible everything about the programs in these countries, and (2) to report on specific topics and refer to the state of the particular art in many different countries. So far we have not attempted the first technique, and we have only occasionally used the second technique, particularly in review articles of conferences.

At the First United Nations International Conference on the Peaceful Uses of Atomic Energy in 1955, nearly one-third of the papers dealt with isotope production and applications, that is, with subjects of interest to DID. In 1958, at the Second Conference, only about one-fifth of the papers were in this same area. At the 1964 Conference the number was down to less than one in fifteen. Ready access to many of these papers may be inconvenient. Yet the articles are of interest to DID and do present a worldwide insight. Hence it seems appropriate to devote an entire issue of *Isotopes and Radiation Technology* to coverage of the 1964 Conference.

Several summaries of wide scope and limited detail have already been published.[1a–1g] These touch only highlights, however, and none has dealt in detail with the specific areas of isotope production and applications covered here.

Because of space limitations it is impossible to include all relevant papers in their entirety. The presentation of those included runs the gamut from almost verbatim copying to annotating titles. Since isotope techniques are more appropriate to this Review than are results, only the salient features from the standpoint of isotope producers and users have been extracted from many articles. In general, peripheral discussions and data not directly related to the subject have been omitted. In three instances (designated by translation numbers), the papers presented in languages other than English have been translated.

References were a serious problem, but the editors believe that they are one of the most valuable features of these papers. With the tremendous number presented, it was impossible to verify the bibliographic data for them all, particularly those in some of the less accessible foreign journals. Those found in the original journals, *Nuclear Science Abstracts*, or *Chemical Abstracts* were corrected as necessary. For the sake of brevity, the titles of articles, conference papers, and reports were omitted from those references which were verified. References not verified are marked with an asterisk.

In the coverage of the Geneva papers, the regular categories of *Isotopes and Radiation Technology* are adhered to. The first two papers are broad overall reviews. The one by Aebersold (USAEC), essentially a follow-up to the review presented in an earlier issue,[2] treats the large-scale production of such isotopes as ^{60}Co, fission products, and transuranics and their applications. It discusses tracer methodology in considerable detail and covers instrumentation. The Russian paper by Gruzin emphasizes the applications of isotopes in physical and chemical research and covers such items as diffusion, the study of alloy structures, radiation damage, and catalysis. The bibliography of this paper is particularly

valuable because of its strong coverage of Russian and other eastern European work.

Several areas of isotope production are covered in Sec. II. Of particular interest to the USAEC is the problem of fission-product separation and recovery. Tomlinson's paper discusses in depth methods and facilities for the recovery of the major fission products in the United States. The report by Brezhneva et al. presents the current status of the Russian extraction procedure for fission-product isolation. The paper by Baetslé et al. and several that follow describe the use of inorganic ion exchangers for fission-product recovery.

The capabilities for reactor production of radioisotopes in several developing countries are discussed in a series of short papers from Brazil, Bulgaria, China, Hungary, Poland, Romania, Spain, Turkey, and the United Arab Republic. Such information is valuable to the USAEC in evaluating its role on a worldwide basis. Several papers on stable-isotope separation are also referenced.

The USAEC Division of Isotopes Development supports a number of programs under the heading of Isotope Technology Development. In Sec. III are papers relating to some of these areas of interest. In *Isotopes and Radiation Technology*, 2(2): 124-33 (Winter 1964–1965), the USAEC program in the field of hydrology was discussed. The paper by Payne et al. (IAEA) reviews this subject on a worldwide basis. It shows the role that isotope techniques are playing in scientific hydrology and emphasizes the benefits to be gained by a wider use of these relatively new methods in such areas as localized and regional groundwater studies, groundwater tracing, borehole logging, studies of surface-water resources (e.g., gaging of river flow, measurement of snow deposits and soil moisture, movement of sand and silt), and pollution studies. Other reports in Sec. III deal with applications of radioisotope tracers in civil engineering (including measurement of the uniformity in ligand spraying and the inclination of strain gages in concrete slabs and, in addition, location of metal parts used to measure the settling of a road surface) and with the estimation of pipeline wear in iron-ore transportation.

Section IV, which deals with process radiation development, contains reviews by Machurek et al. (USAEC) on commercial radiation processing and by Manowitz et al. on chemonuclear processes. Reports from Russia and Poland cover radiation thermal cracking of oil products and a study of the chemical changes in fluids under gamma radiation, respectively.

Facilities of the USAEC, such as Oak Ridge National Laboratory (ORNL) and Brookhaven National Laboratory (BNL), have had long-standing programs in the field of source development. Cless-Bernert and Duftschmid describe a rather novel technique involving radiography with extended sources of radioactive liquids or gases. This paper is reviewed in Sec. V.

The papers in Sec. VI dealing with isotopic power development present recent material in an area previously covered in *Isotopes and Radiation Technology*, 1(1): 89-91 (Fall 1963), and 1(4): 325-39 (Summer 1964). The Martin Company paper is a general discussion of design and technology, and the Russian paper discusses specifically ^{210}Po and ^{144}Ce heat sources.

In the medical-biological-agricultural areas, Belcher et al. (IAEA and WHO) discuss advances in the use of isotopes and radiation sources in medicine. A paper from the United Arab Republic covers the separation and purification of certain medical radioisotopes, one from Brazil discusses tracers in biology, and one from South Africa discusses the use of isotopes to study man's adaptation to environment. A paper on the use of isotopes in agriculture is supported by more than 200 references. Finally, a Brazilian paper relates the effect of liming on phosphate availability in soils as investigated with radioisotopes, and one from the UAR discusses isotopic labeling.

Papers on activation analysis and safety will appear in subsequent issues.

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f. B. Saïtcevsky, *Rev. Franc. de l'Energie*, pp. 53-65 (November 1964).
g. The Third Conference—A Summing Up, *IAEA Bulletin*, 6(4): 7-17 (1964).
2. E. E. Fowler and P. C. Aebersold, *Isotopes Radiation Technol.*, 1(1): 1-26 (Fall 1963).

Advances in Radioisotope Production and Utilization in Science and Industry

By Paul C. Aebersold*

Paper 196

Only three decades after the discovery of artificial radioactivity, man-made radioisotopes have become powerful and versatile tools for a multitude of uses in nearly all fields of research and application. In many instances, discoveries made through the use of radioisotopes are so important and striking that we tend to lose sight of the fact that they would not have been demonstrable but for the availability of a varied supply of radioisotopes. The overthrow of the parity principle in weak interactions,[1] the evolution of a generally accepted theory of Fermi interactions involving beta decay, muon decay, and muon capture,[2,3] and the historic discovery of the Mössbauer effect[4] are magnificent examples.

Although about 130 radioactive species are routinely produced and used, the potential number of useful radioisotopes is much greater. The bulk production of radioactivity is now done in nuclear reactors. Through neutron bombardment in high-flux high-power reactors, production of most useful isotopes can be expanded to meet any conceivable need, up to hundreds of megacuries of isotopes such as ^{60}Co. Also, vast quantities of fission products are obtained as by-products of power or plutonium production. Individual radioisotopes from this source are being separated and purified in megacurie quantities. Cost of production, although greatly reduced, could become even lower with increased demand. Reactor production is supplemented by a variety of accelerators with great flexibility in type and in energy of particles and radiation generated.

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Rapid improvement and expansion in the use of radioisotopes have taken place in all fields. In the basic physical sciences, the major applications are for analytical techniques, determination of molecular structure, measurement of reaction kinetics, and determination of physical properties and behavior of matter. In the applied sciences, new uses are being developed in engineering and in such fields as oceanography, meteorology, hydrology, other geosciences, and space technology.

Advances in radiation detectors, associated electronics technology, and the automatic recording and analysis of data also have been rapid. Use of liquid, plastic, and crystal detectors, with associated circuitry involving multichannel pulse-height analysis and coincidence or anticoincidence techniques, has been extended to extremely low-level counting of samples, even to human beings. Recently a variety of semiconductor detectors has been developed which provide increased energy resolution for counting alpha particles, conversion electrons, and low-energy X and gamma rays. Increasing attention is being given to automatic recording and to computer analysis and reduction of data from isotope and activation analysis procedures.

This paper examines some of the major advances made in isotope production and use in the United States during the 6 years following the Second United Nations Conference on the Peaceful Uses of Atomic Energy.

Large-Scale Production of Isotopes

The production by newly developed processes of extremely large quantities of isolated, purified fission products and neutron products has been the most impressive development in the United States since the last Geneva Conference. The incentive for this effort has come largely from the concurrent development of small electric-power supplies[5] that use the heat of radioactive decay as their energy source.

Of the fission products, ^{90}Sr has received the most attention for isotopic power use. From 1961 to 1964, almost 5 million curies of this isotope were recovered from waste streams and converted to $SrTiO_3$, a ceramic with good thermal and radiation stability, reasonably high strontium content, and relatively low solubility in water.[6]

To date, all nuclear power supplies used in earth satellites have used ^{238}Pu as their energy source. This isotope is produced in kilogram quantities by irradiation of ^{237}Np, which, in turn, is produced by ^{238}U $(n,2n)$ and ^{235}U $(2n,\gamma)$ reactions in reactor fuels.

Cesium-137 is expected to find more use as a gamma-radiation source in radiation processing of foods and chemicals.[7] In anticipation of these uses, a 215,000-curie source of ^{137}Cs was completed in January 1964 by Oak Ridge National Laboratory. The final source, doubly encapsulated in stainless steel, will be used in the radiation development program at Brookhaven National Laboratory.[8] Approximately 1 million curies of ^{137}Cs has been produced by elution of a cesium fraction from an inorganic ion exchanger and purification by the alum crystallization process.[9]

New opportunities for the use of ^{60}Co will arise from a program now under way to manufacture this radioisotope in specific activities of 400 to 500 curies/g or even greater. A capability for the manufacture of hundreds of megacuries per year of ^{60}Co exists in the United States, although total current use is only about 3 Mc. Tritium and ^{14}C are also available in quantities challenging the imagination of those of us who were early workers in the field. Such large reactor capacities for the production of isotopes arise from the increased availability of ideally suited reactor space as a result of the curtailment of plutonium production.

Curium isotopes are receiving much attention at this time for two reasons: they are part of the irradiation chains leading to the very heavy elements, and they appear useful as heat sources in various applications. A facility is being built to process[10] kilogram quantities of ^{244}Cm into targets for further irradiation in neutron fluxes of $>10^{15}$ $n/(cm^2)(sec)$. A special reactor* for this purpose is scheduled for completion in 1965 (Ref. 11). The product of the high-flux irradiation will be gram quantities of ^{252}Cf for study of this element itself and for further irradiation in cyclotrons and other machines. A campaign to provide an initial supply of ^{244}Cm for research purposes was concluded this year. About 400 g of ^{244}Cm was formed by neutron irradiation of 10 kg of ^{239}Pu for several years. This material has been purified and will permit the determination of the macroscopic physical and chemical properties of curium on a much more accurate basis than previously.

The shorter lived isotope ^{242}Cm is also interesting as a heat source. To produce it requires a supply of ^{241}Am, formed by the beta decay of ^{241}Pu. Kilogram quantities of ^{241}Am have been made available for research or isotope production. Although the production of ^{242}Cm by neutron irradiation of ^{241}Am is a straightforward procedure, the subsequent purification of the ^{242}Cm and its fabrication into heat sources pose formidable problems because of the intense heat and alpha radiation as gram quantities of ^{242}Cm are accumulated.

Production of fission-product heat and radiation sources beyond 1968 may require a special plant (Hanford Isotopes Production Plant).[12]

Although the magnitude of radioisotope production in the fission process or by neutron irradiation cannot be approached by machine irradiation, the production of radioisotopes by cyclotron bombardment has nonetheless become increasingly important, particularly for neutron-deficient isotopes. Currently the United States is using 50% of the operating time of a 2.18-m cyclotron[13] originally built for research purposes for the production of radioisotopes. Plans are being made for a more efficient and versatile accelerator for this purpose.

Thermal Applications of Isotopes

One major thermal application of radioisotopes, the use of radioisotopes as heat sources in direct-conversion electric-power supplies, has already been mentioned. As a prediction of things to come, a few other promising thermal applications that are under study in the United States should be mentioned. We are examining the feasibility of using radioisotopes as the energy source in small rocket motors. Three concepts are under consideration: (1) the heat evolved from the decay process raises the temperature of a propellant gas in a direct convective heat-transfer mode, (2) the heat activates a catalyst that accelerates decomposition of hydrazine monopropellant, and (3) radiation heating of the walls of the flow passages is

*This reactor is described in J. A. Swartout et al., *The Oak Ridge High Flux Isotope Reactor*, this Conference, Paper 221. The chemical processing required is summarized in F. L. Culler and R. E. Blanco, *Advances in Aqueous Processing of Power Reactor Fuels*, this Conference, Paper 249. Full details of the chemical processing are in D. E. Ferguson et al., *Nucl. Sci. Eng.*, 17: 435-97 (1963).

used to raise the temperature of the propellant gas. Rocket motors of these types have very high specific impulse but low thrust and would therefore be used in the upper stages of small unmanned space vehicles destined for long trips to other planets or for movement from a low-altitude earth orbit to a high-altitude orbit on a minimum-energy trajectory. Small radioisotope heat sources are also being considered for use in satellites to keep the electronic components at an optimum operating temperature.

Large Sources for Radiation Processing

Another outlet for massive quantities of radioisotopes is expected in the expanding radiation processing industry for the production of plastics and chemicals (Refs. 14, 15, and 17–20), the sterilization of medical supplies,[16] and the preservation of food.[21–24] This work is covered in detail in Paper 198, which is presented in Sec. IV of this review.

Tracer Methodology and Applications

The use of radioactive tracers in every phase of science and industry has become so commonplace that one can no longer be all-inclusive in describing advances in the subject. Other speakers are scheduled to discuss the use of tracers in agriculture, biology, medicine, and hydrology. I will therefore limit my remarks to a few comments on new techniques having rather general utility.

A broad variety of materials can be tagged with ^{85}Kr without regard to the chemical or physical form of the solid substance being labeled.[25,26] This tagging is performed by diffusion of the gas into the material under high temperature and pressure or by bombardment of the material with ionized gas. The krypton gas now contains 5% ^{85}Kr, but, as the technique is developed, higher concentration of the radioisotope will be needed to increase the sensitivity of measurements.

Already some 80 different solids, including elements, alloys, and inorganic and organic compounds, have been impregnated with ^{85}Kr by this versatile method. The amount of the gas absorbed is a function of the physical structure of the material. Pyrolytic graphite, for example, may be labeled with a specific activity up to 1 curie/g, whereas a steel sample will absorb gas only within a few microns of the surface, and the bulk specific activity will be microcuries per gram or less.

Established applications of this technique include the measurement of erosion of turbine blades, wear of bearings, and the kinetic properties of refractory metals at high temperatures.[27] If the krypton is incorporated into a reactive compound, the extent of chemical reaction can be measured by determination of the amount of released radioactivity, a technique similar to the "radio-release" method for ^{14}C and ^{3}H compounds.[28] Devices based on the release of ^{85}Kr are being developed to measure the oxygen content of the atmosphere of Mars and to detect fluorine and hydrogen in manned space vehicles and at launch sites.

At the last Geneva Conference, the total-count method for determining the flow rate of fluids with radioactive tracers was first reported. Since that time the method has found widespread use throughout the world and has become greatly diversified.[29]

The advantages of using noble-gas radioisotopes have been demonstrated by another tracer technique reported recently.[30] Here the 5.27-day ^{133}Xe radioisotope was employed in the conventional dilution method to measure gas flow rates in an organic-chemical plant. Other methods were unsatisfactory because the gases were corrosive and carried entrained particulate matter. The ^{133}Xe was chemically inert, and no corrections for tracer deposition or fractionation were necessary. Flow rates ranging from 50 to 500,000 liters/min were measured with a standard deviation of less than 1%. Counting of gas samples was performed in a large well type plastic scintillator with excellent efficiency.

Measurement of the flow of sulfuric acid through various reactors and circulating loops[31] in a hydrocarbon alkylation plant is an unusually difficult problem. Rapid injection of radiogold into the system and measurement of tracer concentrations in acid samples or tracer flow through pipes permit easy determination of four quantities: acid circulation rates, volume of acid in the system, replacement rate of acid, and entrainment of acid in the hydrocarbon stream.

Tracer methodology has reached the point of prosaic acceptance. A survey of the annual abstracts of national meetings of research societies shows that much work was covered in which radioactive isotopes were used; the

greatest concentration is in the fields of medicine, biology, biological chemistry, and physical chemistry.

Recoil or "hot-atom" chemistry occupies such a significant role in tracer methodology that an entire symposium was devoted to the subject by the International Atomic Energy Agency (IAEA) in 1960 (Ref. 32), and a survey article in 1962 (Ref. 33) on this subject stated that 600 or 700 papers had been published in the field of recoil chemistry since its inception. The survey article itself cited 101 literature references, most of which appeared in 1959 and 1960.

The isotope ^{3}H has found widespread use[34,35] as a radioactive tracer in functional groups of organic compounds. Many workers have used the self-labeling method of Wilzbach,[36] but recently a new technique with substantially broader applications has been reported.[37–40] Hydrogen-tritium exchange is effected by contacting the organic material to be labeled with a tritiated phosphoric acid–boron trifluoride complex.

Raaen et al.[41] have been able to measure the secondary isotope effect in the reaction of acetophenone-β-^{14}C with 2,4-dinitrophenylhydrazine. The ratio of specific rate constants of labeled and unlabeled species proved to be 1.0085 ± 0.0004. By using the same procedure with mixtures of deuterated acetophenone and acetophenone-β-^{14}C, acetophenone-α-^{14}C, and other ^{14}C-labeled ketones, these investigators were able to measure the isotope effect of deuterium substitution for hydrogen in the ketone structures in the reactions that form phenylhydrazones.[42] For the case previously cited, the net effect of deuterating the methyl group in acetophenone was to increase the rate constant by a factor of 1.111. In other words a radioactive tracer was employed to determine the effect of a stable-isotope substitution.

Radioisotopes found in nature are often used as tracers or as chronographs. The classical work of Libby and coworkers employing ^{14}C measurements for dating of archeological samples was honored with a Nobel Prize in 1960. Other naturally occurring isotopes, both stable and radioactive, are being utilized to an even greater extent as the measurement techniques for them become more sophisticated. A major international symposium on dating with radioactivity was held in 1962 (Ref. 43), and several papers have discoursed on the applicability of isotopic measurements or analysis in cosmochemistry, meteoritics, and geochemistry.[44–46]

Notwithstanding the applications for radioactive tracers already developed in science and industry, much remains to be done. In cases where there may be concern over the use of radioactive tracers, such as in large field experiments involving release to the biosphere or into consumer products, "activation" tracing will find expanded use. This involves the use of stable tracers followed by activation analysis to measure the stable-tracer concentrations. Although a beginning has been made in this technique,[47] refinement of the analytical measurements remains the outstanding problem.

Isotope Measurement and Control Systems

Past conferences have dwelt at length on the use of isotopes as sources of radiation in instruments such as thickness gages, density gages, composition gages, and liquid-level indicators,[48] which are now routinely used by industry in many thousands of installations. In some cases competing instruments have been eliminated. We can be certain that use of isotopic measurement and control devices in industry is still in its infancy: *"There seems no limit to the ingenuity of instrument inventors in using the penetration, scattering, or selective absorption of radiations to test materials rapidly and feed back control information required by today's automated system."*[49]

The radiation-scattering principle was extended into a new realm by a device for measuring the density of air at altitudes greater than 40,000 m. This system[50] uses beta-ray forward scattering for sensing the density of air between a ^{85}Kr source and an anthracene scintillation crystal. Background-radiation count rate from an identical detector is subtracted electronically from the primary signal. Sensitivity of both circuits is increased by discriminating against pulses outside an optimum region of the ^{85}Kr beta spectrum. This system has been incorporated into a balloon-borne package that also measures air temperature and pressure by other means, and sequential readings of density, temperature, and pressure are then transmitted to a ground receiving station. A recent calibration flight collected useful data between 21,300 and 37,500 m.

Oceanography is occupying an increasingly greater proportion of the attention of scientists

in the United States today. Again, nuclear instruments are being developed for measurements deep below the surface of the ocean. One of the more complex of these, the Deep Water Isotopic Current Analyzer (DWICA), consists of a circular planar array of 16 scintillation detectors surrounding a centrally located supply of radioactive solution (typically ^{131}I). Periodic injections of solution are swept by the ocean currents to one or more detectors. A computer system analyzes times of arrival of activity at each detector, calculates velocity and direction of flow, and prints out the information in digital form.[51] The DWICA is designed to operate at a maximum depth of 1800 m.

A most exciting recent discovery that will expand the isotope measurement field immeasurably is that of R. L. Mössbauer. Although the Mössbauer effect is of outstanding importance in basic areas of physics, it is also finding other uses. A review of applications of the Mössbauer effect alone would be extremely lengthy. I will hence limit my remarks to a few examples of applications that have appeared recently in the United States.

The most publicized experiment of recent times was the confirmation of Einstein's principle of equivalence by Pound and Rebka,[52] who measured the difference in frequency of gamma quanta emitted at about 20 m above the surface of the earth and at the surface. The difference, as predicted by Einstein, should have been about one part in 10^{16} per meter of altitude, and this was detected quantitatively by measuring the frequency shift of the resonance-absorption peak in ^{57}Fe. In other words the difference in "weight" of gamma quanta at an altitude of 20 m and on the ground was measured! As Goldansky said in a recent review article[53] on the Mössbauer effect, "By comparison with this splendid piece of research, even the most elegant experiments are bound to suffer. . . ."

The greatest application so far has been in the study of the physical and chemical environments of target nuclei whose energy levels are affected by local magnetic fields, charge densities, and electric field gradients. In this sense, we speak of "Mössbauer spectroscopy." By far the favorite target has been ^{57}Fe, with applications ranging from ferrimagnets[54] to hemoglobin.[55] Tin-119 is a favorite target of the Russian workers.

The Mössbauer effect also provides a tool in the study of lattice dynamics and diffusion[56] since the intensity, line shape, and temperature shift are governed by the "stiffness" of the target environment.

Two groups have proposed use of the Mössbauer effect to determine the small relative velocities at which one spacecraft will approach another in a docking or rendezvous maneuver or the closure rate at which a space vehicle will approach a celestial body such as the moon.[57,58] Such velocities should be millimeters per second in the final moments of approach (separation of a few meters). At such time the use of radar is no longer possible, and yet accurate control of speed and alignment is essential to prevent damage from shock or from malfunction of the coupling mechanism.

The use of a Mössbauer source and absorber is probably limited to ranges of 30 m, but at this distance the relative velocities may be in excess of the capability of Mössbauer measurement. Therefore a nuclear gaging system of a more conventional variety, i.e., one that depends on the inverse-square change in count rate with distance from a point gamma-ray source, has been proposed as the intermediate between the Mössbauer sensor and radar.[58]

Instrumentation Improvements

A minor revolution has occurred in the field of radiation detection and analysis since the time of the last Geneva Conference because of two general scientific achievements: the development and commercial production of whole families of solid-state electronic devices or components and the design of very fast data-acquisition equipment and computers.

Crystal scintillators have become more refined and have increased in size. Although thallium-activated sodium iodide remains the workhorse of the field, other inorganic materials, such as cesium iodide and calcium iodide, are receiving increased attention. A recent paper reported[59] that calcium iodide crystals produce light outputs twice as great as those from NaI(Tl) when exposed to gamma radiation and that pulse-height resolutions of 5.2 and 9.9% (full width at half maximum) for ^{137}Cs and ^{57}Co gamma rays have been obtained with this material. However, problems of crystal fabrication remain to be resolved before a widely useful material will be available.

Plastic scintillators up to 40 cm in diameter are now sold in the United States. The Uni-

versity of Miami uses scintillators of this size to measure trace concentrations of radioactive materials in seawater at depths of 10,000 m. In a system of two scintillators and corresponding electronic equipment, ^{60}Co is detected by coincidence counting of cascaded gamma radiation at concentrations as low as 10^{-18} curie/liter in the presence of high background due to the naturally occurring ^{40}K (Ref. 60). Care must be taken, however, to operate at depths sufficient to reduce the cosmic-ray background to negligible levels.

Liquid scintillators have advanced correspondingly, although they, like the plastic varieties, remain inferior to the inorganic crystals in light production and pulse-height resolution. However, they are superior for detecting very low concentrations of gamma emitters or for internal counting of low-energy beta emitters. Several applications for large liquid scintillators have appeared, e.g., a system for detecting less than 1 μc of ^{60}Co moving at a rapid rate past the detector,[61] a detector for measuring tracer levels of radioactivity in chemical process streams,[62] and an apparatus for measuring neutron-proton coincidences produced by antineutrino interactions with hydrogen or deuterium in the scintillator fluid.[63]

The multiplier phototubes used with scintillators are being improved constantly to decrease transit time, increase electron multiplication, and increase electrical stability and life. A tube 60 cm in diameter has been produced.[64] Other tubes are now available with multiplication factors up to 10^8, which permit the detection of single photoelectrons. High-speed counting tubes have recently been reported[65] to exhibit rise times of less than 0.5 nsec (approaching the minimum theoretical value of about 0.2 nsec), transient times of less than 4 nsec, and dark currents of 10^{-9} amp.

The most striking advance, however, has been in the improvement of semiconductor detectors for all types of electromagnetic or particulate radiation. It is now possible to say that semiconductor detectors have come of age, although much remains to be done in improving their stability at ambient temperatures and their efficiency for counting beta and gamma radiation. Silicon and germanium have been studied most. Silicon is superior to germanium in that its noise level at room temperature is low enough to permit it to be used without cooling; germanium normally requires cooling with liquid air to obtain the greatest sensitivity. Germanium, however, is superior to silicon for detection and resolution of high-energy gamma radiation because of its higher atomic number. Both materials can be drifted with lithium ions to increase their resistivity. Sensitive depths up to 1 cm have been achieved in silicon surface-barrier detectors by this technique, leading to detectors capable of better pulse-height resolution than scintillation detectors for gamma radiation of less than 2 Mev. Of course, the counting efficiency of these semiconductor detectors remains in the vicinity of 1% per millimeter of sensitive-region thickness. New semiconductor materials will be investigated within the next few years which will extend the range of gamma energy and increase detection efficiency; cadmium telluride and gallium arsenide appear to be particularly attractive.[66]

Perhaps equally exciting to the nuclear physicist and chemist has been the appearance of advanced electronic equipment to accompany the improved detectors. Transistorized pulse-height analyzers of up to 400 channels are part of the equipment of most of the larger nuclear laboratories in the United States today. For more complex systems in which coincidence, anticoincidence, or time-delay relationships are recorded, instrumentation with up to 20,000 channels of recorded information, each channel having storage of up to 10^5 counts, can be obtained. The subject of multiparameter analysis of this type is receiving great attention since the capacity to record data has now exceeded the capacity to analyze the significance of the data. At a conference held in 1962 in the United States, applications of multiparameter analysis in nuclear physics were discussed.[67]

To the person interested in radioisotopes technology, the techniques of greatest interest are those in which a mixture of gamma spectrums from the simultaneous decay of several isotopes is resolved into the constitutent spectrums to permit quantitative identification of the isotopic mixture or resolution of the complex decay mode of a radioisotope. Such procedures are of utmost importance in neutron-activation analysis, and a number of laboratories are vigorously attacking the problem.[68] The approach being employed is first to assemble a "catalog" of the spectrums of all isotopes of interest in a standard detector geometry and to record the information on punched or magnetic tape by

means of an analog-to-digital converter. An unknown spectrum is then recorded and stored in similar fashion, and a high-speed computer is employed to compare peaks, sort out individual spectrums, and finally calculate the relative amounts of identified isotopes present. The time required to perform the analysis is thus reduced by several orders of magnitude in comparison to hand computations. Excellent summaries of this, and other applications of multiparameter and computer analysis to radioisotope work have been published.[69–73]

In general, the combination of pulse-height analyzers and computer memories, plus fast and graphic readout systems, facilitates the performance of ordinarily laborious tasks, such as calculation of the pulse-height response of a scintillation crystal as a function of gamma-ray energy, computation of coincidence sum spectrums for coincident or cascade gamma emission, resolution of superimposed photopeaks, and angular correlations of coincident events.

The Future of Radioisotope Applications

Notwithstanding the advances made in the last 6 years, our optimism for the future is tempered with the sobering thought that much remains to be done before the potentials of radioisotopes in science and industry are more fully realized. More detailed education of scientists and engineers who are not nuclear specialists is required to acquaint them with the unique characteristics and versatile capabilities of radioisotopes as well as their already proven applications. Scientists and engineers so educated could employ radioisotope technology as a ready tool in the same manner as the more conventional techniques of chemistry, physics, and other basic disciplines.

Regulation of the use of radioisotopes by governmental agencies is sometimes still too restrictive. However, many gains have been made in extending the freedom of use of these materials while maintaining good safety practices. Industrial management has not yet fully recognized that the use of radioisotopes can provide substantial savings in time and money not possible by other means. This requires a continuing program of education at all levels.

Several predictions of technical advances to be realized have been made. We can expect to see many megacurie quantities of specific radioisotopes in use for radiation processing, for small electric-power generators, and for heat sources in outer space. We will see them employed in more routine ways as well as in explorations from the bottom of the sea to outer space—at the extremes of our environment. It has been said before, and it may need to be repeated for many decades, that the applications of isotopes will be limited only by man's ingenuity.

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Use of Isotopes and Radiation Sources in Physical and Chemical Research

By P. L. Gruzin*

Paper 298 (abridged)†

Physics

The use of isotopic and radiation-physical methods of research has led to substantial increases in our understanding of physical phenomenons, e.g., diffusion and distribution of elements in alloys. Melting and solidifying of crystalline bodies, deformation and recrystallization of metals, and stability and weakening of alloys at high temperatures are related to diffusion. Fundamental properties of metals — e.g., plasticity, hardness, stability, and corrosion resistance — and many thermophysical and magnetoelectrical properties of metals and ionic crystals are dependent on the distribution of impurities and structural defects in the lattice.

DIFFUSION

Self-diffusion parameters were first determined with natural radioactive isotopes[1] in 1920 and with artificial ones[2] in 1937. In studies of volume diffusion in solid bodies,[3–13] layers of a flat sample can be removed and the activity of the remainder measured. The values obtained are used to calculate diffusion coefficients.

The change in total activity of a sample can be determined, without destruction of the sample, from the ratio of radioactivities or from a kinetic curve of total radioactivities.[14–16] The ratio of the intensities of the soft to the hard radiations of a radioactive element changes as the element diffuses since the intensity of only the soft radiation decreases. The change in the ratio therefore indicates the extent of the diffusion. The method is simple when the isotope has a beta and a gamma radiation. Changes in dimensions of the sample, vaporization of the isotope, and changes in the geometrical conditions do not have to be considered. The expression for the kinetic curve of total radioactivity can be obtained from values for simultaneous volume and boundary diffusion in a polycrystalline body.[17,18]

Self-diffusion has been studied over a wide temperature range in such important metals as iron, nickel, titanium, zirconium, chromium, molybdenum, and tungsten.[10,19–25]

Results of studies on the diffusion mobilities of alloying elements should be useful in improving the technology of melting and heat-treatment of alloys and semiconductor materials.[26–29]

DISTRIBUTION OF ELEMENTS IN ALLOYS

Contact autoradiographs made with radioactive isotopes or by neutron activation are widely used in alloy-structure studies. Data have been obtained on the effect of allotropy, recrystallization, deformation, alloying, and other factors on the distribution of elements in steels, alloys, and semiconducting materials.[30–32] The microdistribution of impurities and alloying elements in alloys containing zirconium and titanium is of fundamental importance.[33] The heat resistance and corrosion stability of zirconium and titanium alloys depend on the presence of concentration heterogeneities in the microvolumes resulting from the $\beta \rightleftharpoons \gamma$ transformation. Autoradiographic data on segregation of elements in steels and alloys have led to changes in processing technology.[34,35]

Contact autoradiography has been used to study the flow of metal in hot and cold deformation[36,37] and thus to determine the effect of process variables. The results are useful in solving problems of stability of machine parts obtained by deformation and in developing new processes.

Autoradiographic methods have also been used for studying purification of metals in the liquefiable region.[38,39]

Experiments on improving autoradiographic methods[40] have proved that sulfur dispersed in iron is concentrated in the graphite flakes present in the iron. A dendritic macromosaic in aluminum-copper monocrystals and a striated one in aluminum-zirconium crystals have been demonstrated by the autoradiographic method.

Neutron autoradiograms have been obtained which characterize the distribution of small additions of boron in zirconium alloys.

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†Also in *At. Energ. (USSR)*, 17(4): 278–87 (1964); original Russian translated as ORNL-tr-425.

INTERATOMIC ACTION IN METALS

Neutron diffraction has been used to determine partial magnetic moments of nickel and chromium in nickel-chromium alloys,[41] and data have been obtained on the effect of phase composition on ^{119}Sn gamma-resonance-absorption spectrums.

Radioactive-indicator methods have been used to determine, with high precision, very small changes in the concentration of the dissolved elements in a solid solution when an electric current is applied.[42,43] Radioactive isotopes and mass-spectrometer analysis have been used[44,45] to measure the tension of a vapor in equilibrium with the solid phase at different temperatures.

The Mössbauer effect is used for studying defects in metals and the effect of the defects on the crystalline lattice, e.g., the effect on the ^{119}Sn nucleus in matrixes of vanadium, gold, platinum, and thallium.[46] For solvent atoms in tin-base alloys, a dependence of the probability of the effect on the impurity concentration has been discovered; this indicates a change in the force constants in solid solutions.[47] A relation of the effect to melting temperature and atomic volume of a substance has been shown.

NATURE OF RADIATION DAMAGE IN SOLIDS

Methods using polonium-beryllium neutron sources and isotopic gamma equipment have been developed for radiation processing of metals together with methods for internal irradiation of metals with fission fragments.[48]

Data have been obtained on the effects of different types of radiation on elastic and nonelastic properties and on mechanical properties of monocrystalline and polycrystalline samples of copper, zirconium, titanium, chromium, and molybdenum. The effect of gamma and other types of radiation on a Bordoni type internal-friction peak in zirconium and the effect of small gamma doses on the amplitude dependence of internal friction in copper monocrystals, polycrystalline molybdenum, and other metals have been shown.[49] The pair defect of Frankel has been observed after gamma irradiation of metals with 10^8 to 10^9 r (from ^{60}Co).

Work has been done on the effect of nuclear radiation on the course of physicochemical reactions in metals and alloys—in particular, the action of gamma and electron irradiation on aging of copper-beryllium alloys. The interaction of radiation defects with dislocations in steels that have been stabilized by thermomechanical treatment has been studied.

Chemistry

Study of the effect of nuclear radiation on chemical substances and processes is a new field of chemical science—radiation chemistry. Highly efficient radiation-chemical processes can be carried out on an industrial scale.

*RADIATION CHEMISTRY**

Data have been obtained for clarifying the mechanisms of radiation processes in aqueous solutions (Refs. 40, 50–52, 62, and 63). When platinum, palladium, and rhodium complexes in aqueous solutions are reduced by irradiation, a black material forms whose catalytic activity differs from that of catalysts prepared by usual methods. Study of the radiolysis of concentrated solutions has shown that sulfuric acid is decomposed both directly and indirectly by irradiation, but hydrochloric acid, only directly.

Acrylonitrile in the solid phase at −196°C has been shown to polymerize directly at the moment of irradiation without activation energy. This effect may be caused by short-lived active centers that disappear immediately when the beam is switched off.

The initial polymerization rate of tetrafluoroethylene (TFE) in the solid state near the melting point is of higher order than in the liquid state at the same temperature because the activation energy in the crystal is 0.6 to 1.6 kcal/mole, and in the liquid, 6 kcal/mole.[53] By electron paramagnetic resonance (EPR) and thermoluminescence methods, it has been established that paramagnetic and nuclear particles form when solid TFE is irradiated. An ionic mechanism of solid-state polymerization of TFE is indicated.[54]

Polymerization of acetaldehyde, propionaldehyde, and trioxane under the action of X and gamma rays proceeds most effectively in the crystal at 10 to 30°C below the melting point.[55] Polymerization of acrylonitrile, dimethylbutadiene, and cyclohexadiene with urea and thiourea in clathrate type compounds has been studied, and the stereoregularity of the polymers obtained has been established.[72]

*Coauthors of this section are A. D. Abkin, Kh. S. Bagdasar'yan, N. A. Bakh, N. Ya. Buben, V. V. Voevodskii, V. I. Gol'danskii, P. I. Dolin, V. L. Karpov, L. S. Polak, S. Ya. Pshezhetskii, V. L. Tal'roze, N. N. Tunitskii, B. L. Tsetlin, and B. V. Ershler.

The study of mass polymerization at 0°C and 1 to 30 rads/sec showed that, for fluorine and fluorine-substituted compounds of ethylene, G is equal to 10^3 to 10^6. Polymerization of perfluoropropylene in the liquid and solid states gives a low-molecular product with a degree of polymerization no higher than 20, but, at the moment of phase change, the polymerization rate does not change. At doses of 10^7 to 10^9 rads, solid polymers are obtained from various monomers of the allyl series that do not polymerize with ordinary initiators. In contrast to ordinary initiators, irradiation of borazone derivatives gives soluble products with a polymerization degree of about 9 and with borazone rings in the chain. Diphenylvinyl phosphine has been polymerized in the molten state.[56,73,74]

New mineral-organic preparations, made by gas-phase polymerization of monomers, are inorganic sorbents[57] with ion-exchange functional groups. Vinyl monomers have been polymerized, along with olefins and monomers of the acetylene series, on synthetic fibers.[58] The grafted polymer is in the oriented state.

In vulcanization with sulfur combined with heating and irradiation, both polysulfide and carbon-carbon bonds are formed, the ratio of which can be strictly controlled. Molecular destruction is less than that in ordinary sulfur vulcanization or in pure radiation vulcanization. The resins, which contain two types of molecular lattices, are differentiated by higher stability to aging, low hysteresis loss, and high resistance.[75,76]

New data have been obtained on the mechanism of polydimethylsiloxane substitution, and radicals participating in this process have been determined by an EPR method. Radiation vulcanization of different types of rubber in latex has been found to take place with a higher yield than that of rubber in the mass. On simultaneous introduction into the polymer of thermostabilizers and "antirads," synergistic effects have been observed.

In low- and medium-dose studies on radiation cross-linking in mixed polyesters of sebacic and terephthalic acids with ethylene glycol, the extreme member of the series, polyethyleneterephthalate, was destroyed. With high doses it was joined, and with a decrease in the terephthalic acid units, the joining increased.

Dissociation of the adsorbent in a H_2-Al_2O_3 system in a gamma-radiation field was considerable. The adsorption of hydrogen atoms by Al_2O_3 centers depended on the preliminary catalyst activation conditions.[58]

Radiolysis of alkanes adsorbed on various oxide catalysts containing Al_2O_3 took place as a chain reaction above 150°C (Ref. 54). Irradiation (10^8 rads) of cyclohexane sorbed on both insulators and semiconductors showed a series of catalytic activities. With catalysts having properties similar to those of the insulator, paramagnetic centers and adsorbed C_6H_7 radicals were shown by the EPR method.[59] Radiolysis of CO_2 and radiation synthesis of ammonia adsorbed on various oxides (Al_2O_3, SiO_2, ZnO, MgO, NiO, and CuO) which are not catalysts in the absence of radiation but which increase the yield because of energy transfer have been studied.[60]

A suggested mechanism for radiation-energy transfer from a solid body to an adsorbed substance assumes a statistical dispersion of energy in local fields and concentration of it in other "hot" spots on the surface.[61]

USE OF RADIOACTIVE ISOTOPES IN CATALYSIS AND OTHER PROBLEMS

The introduction of ^{35}S into potassium sulfate affects the isotopic-exchange rate of sulfur in the K_2SO_4-SO_3 system at high temperatures.[64] The adsorption of organic dyes on a barium sulfate precipitate containing radium or ^{35}S differs from that on nonradioactive preparations since positive charges are formed on the surface of the $Ba^{35}SO_4$ sample because of the beta radiation, and negative charges on the (Ba, Ra)SO_4 precipitate because of the alpha emission. Adsorption of organic vapor is higher on a radioactive sorbent ($BaSO_4$),[65] and the solubility of chemical compounds changes when radioactive isotopes (cerium oxalate with ^{144}Ce added, yttrium oxide with ^{90}Y or ^{91}Y added, etc.) are introduced.[66,67]

The introduction of radioactive isotopes in many cases increases catalyst efficiency. The dehydration rates of cyclohexanol[68] on magnesium sulfate containing ^{35}S and of isopropyl alcohol[69] on $Ca_3(PO_4)_2 \cdot H_2O$ containing ^{45}Ca or ^{32}P are higher than those with nonradioactive catalysts.

Electron-microscope studies have shown numerous defects and irregularities on the surface of radioactive samples.[70] In the formation of a solid phase from a liquid or gaseous medium, the radiation from radioactive isotopes causes

formation of a large number of new crystallization centers. However, the associated increase in surface area of the radioactive preparations does not explain all the peculiar properties. The continuous formation of electric charges in the samples and on their surfaces plays an important role.

Recently the effect of radiation from radioactive isotopes on electrical properties of metals has been established.[71] Radioactive samples of iron and steel have shown significantly higher corrosion rates in water and moist air than nonradioactive samples of the same metals.

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New Isotopes and Systematics of Nuclides

By I. Selinov*

Paper 583 (abstracted)

When new uses of isotopes are being considered, the possibility of finding new isotopes that may be useful in solving various scientific and practical problems should be discussed.

If a search for new nuclides is to be successful, it is of vital importance to correctly predict half-life and type of decay. Yet, because of insufficient detailed knowledge of atomic nuclear structure, nuclear theory cannot reliably predict half-lives of many unknown nuclides, especially of those of the remote transuranium elements where it is difficult not only to predict the half-life but also to define the region of isotope beta stability.

Therefore this report discusses the principles of construction and the regularities of rational systematics of nuclides and proposes a periodic system of nuclides from which one may predict beta-stable isotopes of transactinides and the half-lives of unknown isotopes of already known elements. From this periodic system of the nuclides, the author draws a number of conclusions about where new isotopes may be discovered and suggests experiments that could be used to find them.

The following is typical of the predictions that are made:

> The map of isotopes still contains many blank spaces and in the near future one may expect with good reason that a great number of new isotopes of both known and unknown elements will be discovered. It is worthwhile to mention that almost all beta-radioactive isotopes which border the region of stable nuclides have already been discovered; therefore one should not expect to discover new stable isotopes. Almost all the beta-stable alpha-radioactive nuclides of known radioactive elements have also been discovered. More than a thousand beta-radioactive nuclides have now been discovered and an even greater number of those isotopes will undoubtedly be discovered in the future.

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Section

II

Isotopes and Radiation Technology

Isotope Production and Development

The majority of the Geneva Conference papers on radioisotope production discussed the separation of fission products from reactor-irradiated uranium. These papers are presented in the first part of this section. A few countries gave status reports on their isotopes-production programs, usually in connection with the operation of a research reactor. These papers are summarized in the second part of this section. Also included is a list of some papers on research and testing reactors that can be used in radioisotope production. The third part of the section is an annotated list of papers covering the separation of stable isotopes.

FISSION-PRODUCT RECOVERY

This section presents the papers on fission-product recovery methods given at the Geneva Conference. The first paper summarizes the current status of fission-product production in the United States, and this is followed by a Russian paper that gives some of the current processes for fission-product recovery in that country. Papers from Belgium, France, Austria, and Sweden, dealing mainly with the recovery of fission products by ion exchange, are then presented in condensed form. The section is concluded with abstracts of three papers that discuss specific fundamental properties of fission products: two - Russian papers, one on solvent extraction with phosphorus-containing acids and the other on the chemistry of technetium, and a United Arab Republic paper on ruthenium complexes.

Large-Scale Fission-Product Recovery

By R. E. Tomlinson, B. F. Judson, and L. L. Zahn*

Paper 252

Since 1946 Oak Ridge National Laboratory (ORNL) has been isolating and purifying radioisotopes and making them available to other organizations for development and industrial purposes.[1] Through December 1963, 2.4 Mc of more than 100 different isotope preparations have been shipped. Recently the demand for a few fission products has increased sharply, and the recovery of these isotopes has started at other sites. This paper summarizes technologies for the isolation and purification of these isotopes, describes briefly the large-scale production activities, and indicates plans for providing increased amounts of some selected isotopes.

Process Technology

Major strides have been made during the last 6 years in the development of processes for the isolation and purification of four fission products: ^{90}Sr, ^{137}Cs, ^{144}Ce, and ^{147}Pm. Technologies have also been developed for the recovery of other isotopes in which presently there is less interest, e.g., ^{85}Kr, ^{133}Xe, ^{95}Zr-^{95}Nb, and ^{99}Tc. Most early recovery methods were based on selective precipitation techniques; more recently processes have emphasized the use of solvent extraction and ion exchange. Major technical advances include the

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discovery of selective extractants for cesium, the development of organic phosphates as cation extractants, and the use of aqueous-phase complexants to minimize the extraction of undesired cations.

STRONTIUM, CERIUM, PROMETHIUM, AND ZIRCONIUM

Some of the processes available for the recovery of ^{90}Sr, ^{144}Ce, and ^{147}Pm are summarized in Table 252-1. These three fission products can be coextracted from Purex acid wastes and subsequently separated for purification. Strontium-90 can also be recovered from the precipitated salts in stored alkaline wastes.

Table 252-1 PROCESSES FOR RECOVERING STRONTIUM, CERIUM, AND PROMETHIUM FROM ACID MEDIA

Operation	Method*	Reagent	Status†	References	Remarks
Sr, Ce, RE removal from purex acid waste	PPT	Sulfate	P	2	$PbSO_4$ carrier; metathesize with Na_2CO_3
	SE	D2EHPA	P	3, 4, 6, 17, 18	Extract at *p*H 4 to 5
Sr separation from Ce and rare earths	PPT	Oxalate	P	2	
	SE	D2EHPA	P	3, 4, 6, 17, 18	Selectively strip Sr from solvent
	IX	Cation exchanger	L	27	Use to purify Sr
Ce separation from rare earths	SE	D2EHPA	D	3, 5, 6	Selectively strip rare earths (including Pm) leaving Ce(IV) in solvent
	PPT	Peroxyacetate	D	28	Precipitate at *p*H 4
Pm separation from rare earths	SE	TBP	L	8, 9	Extract with 100% TBP
	SE	D2EHPA	L	10	
	IX	Cation exchanger	L	11, 29	Chromatographically elute, using chelating and restraining ions

*PPT denotes precipitation; SE, solvent extraction; IX, ion exchange.
†P denotes extensive use in production activities; D, demonstration in production-plant equipment; L, demonstration in laboratory equipment.

Precipitation with $PbSO_4$ as a carrier for the radionuclides and tartaric or glycolic acid as a complexant for iron and other metals is now being used to separate ^{90}Sr, ^{144}Ce, and rare earths from Purex acid wastes.[2] Strontium-90 is separated from ^{144}Ce and the rare earths by carbonate metathesis and dissolution in HNO_3, followed by oxalate precipitation. Product recovery averages about 70%; less than 5% of the other fission products remain with the products.

Solvent extraction with di-2-ethylhexyl phosphoric acid (D2EHPA) phase-stabilized with tributyl phosphate (TBP) in a kerosene type diluent is a highly versatile process for isolating and purifying ^{90}Sr, ^{144}Ce, rare earths, and ^{95}Zr-^{95}Nb (Ref. 3). The ^{90}Sr, ^{144}Ce, and rare earths can be coextracted from Purex acid wastes, and the ^{90}Sr can be selectively stripped into dilute HNO_3. Thus a relatively pure cerium–rare-earth fraction is left to be stripped into a stronger acid solution.[4] If ^{144}Ce is oxidized with permanganate or silver-catalyzed persulfate and coextracted into D2EHPA with rare earths, the rare earths can then be selectively stripped into dilute HNO_3, and the tetravalent cerium will be left in the solvent.[5] Alternatively, a complexant, such as ethylenediaminetetraacetic acid (EDTA) or citric acid, can be used to suppress the extraction of iron, aluminum, ^{144}Ce, and rare earths and thus permit ^{90}Sr to be selectively removed from Purex acid waste.[6] By similar techniques, separated ^{90}Sr can be further freed of calcium[7] and other ions. These techniques have produced ^{90}Sr containing less than 5 wt.% of metallic impurities and less than 10^{-4}% (by beta emission) of radioisotopes other than ^{90}Sr (Ref. 6).

Promethium-147 can be separated from the other trivalent rare earths by solvent extraction with pure TBP[8,9] or D2EHPA.[10] Chromatographic ion exchange with organic cation resins is also effective in fractionizing the rare earths.[11] *N*-hydroxyethylenediaminetriacetic acid, EDTA, and diethylenetriaminepentaacetic acid (DTPA) are effective as chelating agents for progressive elution; inert yttrium, aluminum, zinc, or hydrogen can be used as a band-restraining ion. Product purities greater than 99% ^{147}Pm have been obtained.

CESIUM

Several processes for separating ^{137}Cs from radioactive wastes are summarized in Table 252-2. Some of the natural and synthetic zeolites, particularly the aluminosilicates, selectively adsorb ^{137}Cs from either acid or alkaline solutions.[12] A synthetic zeolite is now being used to extract ^{137}Cs with high yield from aged Purex waste. The ^{137}Cs is eluted from the zeolite with ammonium ion; if ammonium carbonate is used, the elutriant can be destroyed by heating the solution, and a relatively pure solution of Cs_2CO_3 is left.[13] Cesium recoveries of 99% and a ^{137}Cs/Na decontamination factor of 10^3 are obtained in a single cycle.

Selective precipitants, such as nickel ferrocyanide[14] and phosphotungstic acid,[15] have been successfully tested in production-plant equipment for isolating ^{137}Cs from alkaline and acid wastes, respectively, with yields greater than 95%. Fractional crystallization of cesium aluminum sulfate, which is used to purify and concentrate ^{137}Cs (Ref. 1), gives a very pure cesium product in high yield.

Substituted phenols, such as 4-*sec*-butyl-2-(α-methylbenzyl) phenol (BAMBP), show considerable promise as solvent extractants for separating ^{137}Cs from alkaline solutions; ^{137}Cs is readily stripped from the solvent with dilute HNO_3 (Ref. 16). Cesium recoveries greater than 99% with ^{137}Cs/Na decontamination factors of several thousand are obtained in a single cycle.[3] When BAMBP is combined with D2EHPA, a synergistic effect permits the coextraction of ^{137}Cs with ^{90}Sr and the rare earths from slightly acid solutions.[17] BAMBP is degraded by HNO_2, however, and its usefulness may be limited in highly radioactive nitrate solutions.[18]

TECHNETIUM

As shown in Table 252-2, technetium separation from a UF_6 stream has been effected on a routine production basis by sorption on MgF_2 (Ref. 19). From alkaline Purex wastes, ^{99}Tc can be readily adsorbed on a strong-base anion-exchange resin;[20] ruthenium was the only significant contaminant in a kilogram-scale plant test. Technetium can be extracted from strong acid solutions with tertiary amines as the solvent[21] and purified by either anion-exchange or solvent-extraction techniques.

KRYPTON AND XENON

A capability for recovering rare gases from irradiated fuels has been demonstrated on a large scale. Initially, rare gases were isolated at the Idaho Chemical Processing Plant (ICPP) by low-temperature adsorption on charcoal beds and purified by selective adsorption-desorption.[22] Subsequently, a low-temperature fractional-distillation process was demonstrated to be capable of providing adequate recovery and purification efficiency for routine production use.

Production Activities

Facilities to separate and purify selected fission products have been installed at ICPP, Paducah, Ky., and Hanford to assist the Oak

Table 252-2 PROCESSES FOR RECOVERING CESIUM AND TECHNETIUM

Source*	Method†	Reagent	Status‡	References	Remarks
		Cesium Recovery			
Alkaline wastes	IX	Zeolite	P	12, 13	Elute with NH_4NO_3 or $(NH_4)_2CO_3$
Alkaline wastes	PPT	Nickel ferrocyanide	D	4, 30, 31	Metathesize with silver
Alkaline wastes	SE	Substituted phenols	L	16, 32	Strip with dilute HNO_3
Alkaline wastes	SE	Dipicrylamine	L	33, 34	Strip with dilute HNO_3
Acid wastes	PPT	Aluminum sulfate	P	1	Fractional crystallization
Acid wastes	PPT	Phosphotungstate	D	15	Metathesize with NaOH
Acid wastes	SE	BAMBP-D2EHPA	L	17, 18	Synergistic effect at *p*H 4 to 5
Acid wastes	IX	Zeolite	L	12	Scrub Zr-Nb with oxalate
		Technetium Recovery			
Gaseous UF_6	IX	MgF_2	P	19	Strip with water
Acid solutions	SE	Tertiary amine	P	21	Strip with NaOH or Na_2CO_3
Alkaline solutions	IX	Anion exchange	P	20	Elute with concentrated acid

*Alkaline wastes denote the supernatant solutions, free from precipitated salts, in the stored wastes; acid wastes include the salts of process chemicals and corrosion products.

†PPT denotes precipitation; SE, solvent extraction; IX, ion exchange.

‡P denotes extensive use in production activities; D, demonstration in production-plant equipment; L, demonstration in laboratory equipment.

Ridge Fission Product Development Laboratory in supplying established and projected markets.

BARIUM-140 AND LANTHANUM-140

Recovery of ^{140}Ba-^{140}La from irradiated uranium in the ICPP was started in 1956. In the separations process, ^{140}Ba and strontium were precipitated as nitrates by concentrating an HNO_3 solution of irradiated uranium.[22] The precipitated salts were dissolved in water, the *p*H was adjusted to 4.25 ± 0.25, and the barium was reprecipitated as $BaCrO_4$. Additional dissolution and precipitation cycles provided the required purification. About 2 Mc of ^{140}Ba has been produced in this way.

TECHNETIUM-99

In 1960 the recovery of ^{99}Tc was begun at the gaseous diffusion plant in Paducah. More than 25 kg of ^{99}Tc has been recovered to date. Initially, the ^{99}Tc was collected by washing residue from UF_6 cylinders with water after the UF_6 had been transferred into the diffusion cascade. Later, technetium was isolated by sorption from UF_6 on MgF_2. The ^{99}Tc was concentrated and purified by an anion-exchange process, followed at Oak Ridge by solvent extraction with a pyridine derivative.[22a]

STRONTIUM-90

In 1960 the separation of ^{90}Sr from Purex wastes was initiated at Hanford using the precipitation techniques described above. The separated strontium was concentrated and stored to permit decay of ^{89}Sr. In 1961 an available pilot plant was reactivated to purify the ^{90}Sr by solvent extraction with D2EHPA as the solvent.[6] The purified ^{90}Sr was precipitated as the carbonate for transfer to other sites, where the ^{90}Sr was compounded and stored or encapsulated for use. Through December 1963 more than 6 Mc of purified ^{90}Sr was produced.

CESIUM-137

Separation of ^{137}Cs from the supernatant solution stored in underground waste tanks was begun at Hanford in 1961. The aged waste solution was clarified and passed through beds of a pelleted zeolite, which preferentially adsorbed the ^{137}Cs; the other waste constituents were returned to underground storage. Through December 1963, 1.5 Mc of ^{137}Cs was adsorbed and transferred to Oak Ridge, where the cesium is desorbed and purified.

PROMETHIUM-147

Since 1963 a rare-earth fraction has been isolated from Purex wastes at Hanford using the precipitation techniques described above. The concentrate is stored as an acid solution. Through December 1963 an inventory of rare-earth salts containing 5.2 Mc of ^{147}Pm had been accumulated for aging. Kilocurie quantities of purified ^{147}Pm have been produced at Oak Ridge and at Hanford.

CERIUM-144

In 1964 nearly 0.5 Mc of ^{144}Ce was isolated in the Hanford Purex Plant using the carrier precipitation and purification techniques described above. The ^{144}Ce was precipitated as the sulfate for shipment to Oak Ridge. Earlier, kilocurie quantities of highly purified ^{144}Ce were produced at Oak Ridge.

Intersite Transfer

Radioactive materials are sometimes transferred to other sites so that optimum use can be made of processing facilities. When these transfers are made, the protection of the public is of primary concern. Ionizing radiations must be attenuated to safe levels, and the radioactive materials must be assuredly confined within the shipping container, even under such unplanned conditions as severe impact or fire. Characteristics of three types of transport facilities now in use are described in Table 252-3.

SHIELDED TRANSFER TANK

Several shielded transfer tanks, formerly used to transport aged fission-product wastes,* have been modified to transport ^{137}Cs. The tanks are filled with pellets of a synthetic zeolite on which the ^{137}Cs is adsorbed. After being flushed with demineralized water, the vessel is sealed with three independent closures in series. For intersite transfer, up to three such tanks are fastened securely in a gondola type railroad car,† which has been strengthened by heavy structural-steel members extending over the tanks to protect them from accidental crushing. Cylindrical steel fire shields protect the tanks from direct impingement of flames.

*See *Isotopes Radiation Technol.*, 1(1): 36-8 (Fall 1963).

Table 252-3 CHARACTERISTICS OF SHIPPING FACILITIES FOR FISSION PRODUCTS

	Shielded transfer tank	Solution carrier	HAPO-IB	HAPO-II
Product form	Cs adsorbed on zeolite	$Pm(NO_3)_3$ solution	Dry $SrCO_3$, dry $Ce_2(SO_4)_3$*	Dry $SrCO_3$
Product container	Tank	Tank	Annular filter	Annular filter
Size:				
Diameter, cm	152	29.2	44.0	24.2
Thickness, cm	–	–	1.6	1.1
Volume, liters	1950	19.5	12.0	5.9
Rated capacity:				
Isotope, kilocuries	^{137}Cs, 50	^{147}Pm, 100	^{90}Sr,† 500 ^{144}Ce, 1000	^{90}Sr,† 200
Limitation	Volume	Volume	Volume	Heat dissipation
Performance rating:				
Internal pressure, atm	6.7	43.5	13.3	13.3
At temperature, °C	150	260	540	540
Heat-dissipation capability, watts	515	290	10,000	3400
Shielding:				
Metal	Pb	U	Pb	Pb
Thickness, cm	9	12.7	22.2	16.5
Weight, metric tons	17.2	3.0	18.2‡	8.2‡

*Actually, the double salt cerous sodium sulfate is the precipitate used.

†Limitation also permits 1250 kilocuries of ^{89}Sr to be present in the HAPO-IB carrier and 500 kilocuries in the HAPO-II carrier.

‡The cask container weighs an additional 16.5 and 10.5 metric tons for the HAPO-IB and HAPO-II units, respectively.

SOLUTION CARRIER

An extremely rugged solution carrier for shipping small volumes of high-activity solutions has been used for promethium nitrate solution. The product-containing vessel is fastened within the cask cavity by bolts; liquid mercury provides a thermal bond between the product vessel and the outer shield. Three closures are made in series to avoid any possible leakage through the process connections. The solution carrier is so compact and rugged that mechanical failure would not occur under credible conditions of impact. A fire shield protects the cask from any direct flame, and intumescent paint on the fire shield protects the cask from potential reradiation of heat from the fire shield.

HAPO CASKS

The HAPO transfer systems[23,24] are characteristically dry; a precipitated fission-product salt is collected and dried within the cask for shipment. Each shipping system consists of three main parts: a product container, a cask, and a cask container.

The product container is an annular tank designed to collect and contain the product in the geometry required for safe heat dissipation. The tank contains a filter screen supported by lands created by a series of vertical and horizontal grooves milled into the inner tank wall. The container is loaded by transferring an aqueous slurry of the product salt into the product cavity; the filtrate passes through the filter screen and out of the tank through the vertical grooves in the tank wall. The product salt is dried by heating the unit to about 260°C and purging it with helium. At the receiving site the product is removed from the container by dissolving the salt in a suitable medium.

The cask, which provides radiation shielding and an extended surface for dissipating heat, is designed to withstand a force of 50 *g* without failure. The product container is attached to the cask cover with comparable mechanical integrity. The heat of radioactive decay is transferred from the surface of the product container to the cask by convection through a low-melting alloy.

The cask container is designed to reduce and distribute any decelerating forces that might be encountered and to permit the airflow required for dissipating heat. Four concentric cylindrical steel shells are capped by heavy steel members which are solidly attached to the outer shell and strengthen the unit against end, side, or edge impacts. The shells and end plates are separated by short rubber columns that provide a minimum distance of about 40 cm in which the cask could decelerate under impact conditions. Experimental data confirm that forces on

the cask would not exceed 50 *g* if the cask container were to strike an unyielding surface at a velocity of 13 m/sec (~30 mph).

ADMINISTRATIVE PRECAUTIONS

Although transport facilities were designed to permit the intersite transfer of fission products without need for restrictive shipping procedures, administrative precautions are taken to minimize the possibility of damage to the shipment. These precautions include the choice of train routings to avoid heavy traffic in large population centers and to avoid areas in which large amounts of flammable or explosive liquids and chemicals are stored. A courier trained in emergency protective measures accompanies each shipment.

Production Plans

Plans are now being implemented to increase the rate at which selected fission products can be recovered.[25,26] [Current plans for the recovery of fission products at Hanford were reviewed in *Isotopes and Radiation Technology*, 2(2): 99-123 (Winter 1964–1965).]

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Isolation of Fission-Product Radioactive Elements by Extraction

By N. E. Brezhneva, G. V. Korpusov, E. N. Patrusheva, N. P. Prokhorova, and Yu. S. Krylov*

Paper 512

Since the Second Geneva Conference, long-lived radioactive fission products, such as ^{90}Sr,

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^{147}Pm, ^{144}Ce, and ^{137}Cs, have been increasingly needed. Thus the need for developing simple technological techniques, such as solvent extraction, for separating fission products has grown sharply.

Ten years ago extraction by organic solvents was applied only to the separation of uranium and plutonium from fission products and from each other. In some cases organic solvents have been recommended for separating Ce(IV). At the Second Geneva Conference, a combined extraction-precipitation scheme for separating radioactive fragment elements was reported by the USSR.[1] At Copenhagen in 1960 the United States[2] reported on an extraction scheme for separating fission products.

Published data[3–7] on regularities of extraction behavior of elements now permit development of a combined extraction scheme for separating radiochemically and chemically pure fission products. This report deals with the fundamental questions of isolation of long-lived ^{90}Sr, ^{144}Ce, ^{147}Pm, and ^{155}Eu by extraction.

Basic Regularities in Extraction of Fission Products

The four long-lived radioactive fission products ^{90}Sr, ^{144}Ce, ^{147}Pm, and ^{155}Eu are in the alkaline-earth and rare-earth groups of elements. Since cerium and europium can be easily oxidized or reduced, the extraction process can be divided into four steps: (1) extraction of tetravalent cerium, (2) extraction of trivalent rare earths, (3) extraction of divalent europium, and (4) extraction of alkaline earths.

EXTRACTION OF ^{144}Ce

In the extraction of cerium, as in all methods of its isolation, it is first oxidized to the tetravalent state either chemically, e.g., by $KBrO_3$, $KMnO_4$, $KClO_3$, $NaBiO_3$, or O_3, or electrolytically. Oxidation by $KBrO_3$ or O_3 is performed in $7M$ to $8M$ HNO_3; in more dilute solution, oxidation is incomplete owing to a decrease in oxidation potential. The use of $KMnO_4$ is complicated by the fact that Mn(IV) may form at the same time and, being extractable, contaminate the cerium product. The manganese may be removed from the organic phase by washing with citric acid, or it may be reduced with oxalic acid.

In contrast to trivalent rare earths, tetravalent cerium is easily extracted by many oxygen-containing organic solvents and therefore can easily be separated from other rare earths. At present, tributyl phosphate (TBP) and di-2-ethylhexyl phosphoric acid (D2EHPA) are generally used for cerium extraction.

By TBP. The reaction of Ce(IV) during extraction by TBP from nitrate solutions can be represented by

$$Ce^{4+}_{aq.} + 4NO^{-}_{3aq.} + n(H^{+} + NO^{-}_{3})_{aq.} + TBP_{org.} \rightleftharpoons H_n[Ce(NO_3)_{n+2}]\cdot 2TBP_{org.}$$

where n is less than or equal to 2 in accordance with the cerium coordination number of 6. This equation shows that, depending on the HNO_3 concentration in the aqueous phase, Ce(IV) can be extracted either as the nitrate or, at higher acidity, as a complex acid.

It has been shown, both by saturating TBP with cerium nitrate solution and by physical and chemical methods, that Ce(IV) is extracted as a dibasic complex acid with two TBP molecules, in contrast to the trivalent rare earths, which are extracted with three TBP molecules. Thus, in the purification of Ce(IV) by dilute TBP, as the Ce(IV) concentration decreases, the cerium distribution coefficient decreases as a quadratic function while those of the trivalent rare earths decrease as a cubic function. Diluents used for TBP are carbon tetrachloride, toluene, xylene, and various kerosene fractions. However, when kerosene is used as a diluent at high cerium concentrations, the organic phase divides into two phases of different composition; with other diluents, no such complications arise.

Since cerium is extracted by TBP much more readily than trivalent rare earths under the same conditions, it displaces them from the organic phase. For example, the distribution coefficient for europium from $4M$ HNO_3 decreases from 0.50 to 0.03 when the organic phase is saturated with Ce(IV).

The experimental data indicate that ^{144}Ce should be extracted by TBP from $4M$ to $5M$ HNO_3. With $4M$ HNO_3 and 100% TBP at an aqueous to organic ratio of 5 to 1, 98% of the ^{144}Ce goes into the organic phase in a single extraction, accompanied by 10% of the rare earths. Additional washing is required to purify the ^{144}Ce. With 50% TBP, 90% of the ^{144}Ce but only 1% of the rare earths are extracted in a single

extraction, and pure product is obtained without additional washing.

By D2EHPA. Cerium(IV) extraction by D2EHPA proceeds mainly by cationic exchange between a cerium ion and the acidic hydrogen of the extractant:

$$Ce^{4+}_{aq.} + 4(H_2A_2)_{org.} \rightleftharpoons [Ce(HA_2)_4]_{org.} + 4H^+_{aq.}$$

where H_2A_2 is the extractant dimer molecule. This extraction mechanism is valid with low HNO_3 concentrations in aqueous phase. When extraction is from high-acid solutions, the extraction is brought about by solvation with extractant molecules through the phosphoryl oxygen. In accordance with the extraction mechanisms shown, Ce(IV) distribution coefficients decrease as the HNO_3 concentration in the aqueous phase increases.

In contrast to Ce(IV), trivalent rare earths are extracted by D2EHPA with very low distribution coefficients over the entire range of HNO_3 concentrations except very low ones. Hence cerium should be extracted from a 2*M* HNO_3 or lower aqueous phase. As the Ce(IV) concentration in the organic phase is increased, trivalent rare earths are displaced since their distribution coefficients decrease with increasing cerium concentration in the organic phase.

EXTRACTION OF TRIVALENT RARE EARTHS

Rare earths constitute 56% of all fission products. It is difficult to obtain chemically and radiochemically pure ^{147}Pm and ^{155}Eu because of the similarity of their chemical properties. All separation methods for rare earths are based on the differences in stabilities of their complex compounds, hydrates, or solvates. The maximum difference in stability constants of various compounds of two neighboring rare earths is 2 to 3; the difference in their distribution coefficients is also 2 to 3.

Of several hundred extractants tested for separating rare earths, neutral and acid organophosphorus compounds, e.g., TBP, di-2-isoamyl methyl phosphonate (DiAMP), and D2EHPA, find the widest application. Aliphatic carboxylic acids (7 to 9 C fraction) are very cheap and may also be used.

The presence of the functional group P=O is characteristic of neutral organophosphorus extractants. Extraction takes place by formation of bonds between phosphoryl oxygen and the metal or hydrogen of the rare-earth complex acid. At low or moderate acidity, compounds of the form $M(NO_3)_3 \cdot 3S$ are extracted (S is an extractant molecule); at high acidities, rare earths are present in the organic phase as complex acids, with the limiting composition $H_3M(NO_3)_6 \cdot 3S$.

The highest distribution coefficients in TBP extraction are 1.8 to 2 for high-acid solutions. Although these values are lower than those with dialkyl phosphoric acids, TBP has the advantage that with it the distribution coefficients of the rare earths are essentially independent of their concentrations over a wide range. This is very important since, after the concentration step that precedes separation, the rare-earth concentration may be 40 to 50 g/liter. Results are similar in extraction by DiAMP, for which distribution coefficients are somewhat higher.

Systems suitable for concentrating rare earths are (1) TBP or DiAMP with a salting-out agent in weak-acid solution and (2) phosphine oxide with a slightly acid solution.

Use of Complexing Agents. When inversion occurs or rare earths are extracted with practically no separation, complexing agents can be introduced into the system to obtain maximum selectivity. These agents and their compounds with rare earths are soluble only in aqueous solution. Under inversion conditions the heavier rare earths, which complex more readily in the aqueous phase, are extracted less satisfactorily than the lighter ones. For example, in the nitrilotriacetic acid–carboxylic acid system, which is a poorly selective extractant, the following distribution coefficients have been obtained: La-Ce, 6; Ce-Pr, 3; and Nd-Pr, 2.2.

In TBP extraction the distribution coefficients of the rare earths [Nd, Pr, Ce(III)] are changed very little by increasing their concentration to 50 to 100 g/liter. When acid extractants are used or complexing agents are introduced, one phase should be completely saturated. Then the extraction separation is based on mutual displacement of the elements, and separation values are fairly constant.

DIVALENT EUROPIUM

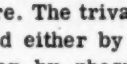
Almost all methods of europium separation are based on its reduction to the divalent state or to a metal. In the case of the divalent salt, europium is precipitated or coprecipitated as the sulfate; in the case of the metal, an amal-

gam is formed. Recently europium has been separated by extraction after preliminary reduction to the divalent state. The europium is reduced by zinc dust or zinc amalgam or at a mercury cathode. Since europium has a low oxidation-reduction potential (0.43 volt), it is readily oxidized by atmospheric oxygen. Therefore the extraction should be carried out under an inert atmosphere. The trivalent rare earths have been extracted either by D2EHPA from a chloride medium or by phosphonates or triphosphoxides from a nitrate medium.

STRONTIUM SEPARATION

Present extraction methods for separation are applicable to practically all metals of the periodic system, but the greatest difficulties are encountered for alkaline-earth and alkali metals. The most serious work on separating alkaline-earth elements, aimed at obtaining pure ^{90}Sr, was reported in Copenhagen in 1960.

We tested many complexing agents and extractants for the separation of alkaline-earth elements, and the most suitable was a solution of salicylaldoxime (SAO) in TBP-ammonia with an aqueous solution of alkaline earths.

Salicylaldoxime,  , has the same active groupings as oxyquinoline derivatives. The distribution coefficient of SAO itself ($\gg 80$) does not change significantly as the *p*H changes from 0 to 12.

The distribution coefficients of the alkaline earths are high only when organic compounds such as TBP are diluted. Equilibrium is reached within several minutes; the complex-forming reaction is reversible.

As the concentration of the complexing agent changes, the distribution coefficients of calcium, strontium, and barium change directly with change in the SAO concentration, which indicates dimerization of SAO in the organic phase. The distribution coefficients for the calcium–strontium and strontium–barium pairs (ammonia concentration 0.1*M* to 2*M*) average 80–50 and 2–3, respectively. With these distribution coefficients the separation of calcium from strontium is not difficult and may be done in a batch or continuous process with a minimum number of stages.

Extraction Installations

There are various practical extraction processes for radioactive isotope separation. In the case of europium, strontium, and cerium, a batch extraction is adequate. Multiple-stage extraction installations adapted for remote control should be used in the separation and concentration of the beta- and gamma-emitting trivalent rare earths (^{155}Eu and ^{147}Pm).

Two types of extractors, countercurrent and semicountercurrent, are used to separate the rare earths. Both types are mixer-settlers. Since the extraction installations are intended for remote operation, mixing is accomplished by air bubbling or shaking. To save installation space, the extraction vessels are tubular.

The vessel has the following features: (1) the settling zone is a horizontal tube connected to a vertical separator; this feature increases the efficiency. (2) The separator has a water seal at the outlet of the heavy phase; this arrangement stabilizes the position of the phase interface over a range of densities.

The total throughput of the countercurrent unit is rated at 20 to 30 liters/hr and allows a variation in the stream ratio from 1/10 to 10/1. Air consumption is about 150 liters/hr per vessel. Regulators having a special device to prevent their contamination are placed on each vessel to control the air usage during operation. Vessel efficiency is close to 100%. A plant of 30 extraction units ensures a pure promethium product.

The semicountercurrent extractor consists of separate vessels; one phase in each vessel is stationary, and the other passes successively through all the vessels. The semicountercurrent plant of 50 extraction units is also intended for remote control. The settling chamber is horizontal. The emulsion is circulated spirally through the settlers, as well as between the vessels, by an airlift. The vessel operates stably with a phase ratio in the emulsion of 1/10. Special safety devices in the vessels ensure automatic switching off of any vessel that stops operating.

Process for Separation of Long-Lived Fission Elements

The initial solution to be separated is a nitrate concentrate of fission products containing corrosion products and impurities introduced into the solution from reagents. The first operation, concentration of strontium and rare earths, is performed by oxalate coprecipitation

of rare earths and alkaline earths. The nitrate solution of rare earths and strontium obtained by destruction of the oxalic acid contains practically no iron, chromium, or other radioactive or stable impurities. The total content of rare earths in the final solution is 5 to 10 g/liter; the radioactivity of the solution depends on its age.

The first stage is (1) extraction of ^{144}Ce by 0.5*N* D2EHPA or 50% TBP after oxidation with ozone or bromate in 7*M* to 8*M* HNO_3 or (2) extraction of all rare earths by D2EHPA from a weak-acid or buffer solution followed by washing of the organic phase with the oxidant in HNO_3 solution so that ^{144}Ce remains in the organic phase and trivalent rare earths go into the aqueous phase. When the ratios of the volumes of initial solution, extractant, and washing solutions are chosen properly, the initial concentrate of radioisotopes is separated into three fractions: pure radioactive ^{144}Ce; ^{90}Sr concentrate; and ^{147}Pm concentrate with other rare earths but no ^{144}Ce.

In all cases, pure ^{90}Sr is obtained after isolation of ^{144}Ce and separation of rare earths. The main impurities from which ^{90}Sr must be separated are calcium, which may be 20 to 30 times the weight of ^{90}Sr, and some barium and lead. Calcium is removed by extraction with 1*M* to 2*M* SAO solution in TBP from an approximately 1*M* ammonia solution of alkaline earths. About 99% of the calcium is extracted and more than 85% of the strontium is left by successive extraction, with three equal volumes of a 2*M* solution of SAO in TBP, from 1*M* aqueous solution, which contains NH_4NO_3, $NaNO_3$, 0.4*M* $Ca(NO_3)_2$, and 0.005*M* $Sr(NO_3)_2$.

Pure ^{147}Pm is produced by TBP extraction, either in the countercurrent installation only or by combination of countercurrent and semicountercurrent processes.

The radioactive europium is separated by extraction after reduction with zinc dust or with zinc amalgam in a chloride medium.

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Inorganic Ion Exchangers in Acid Medium for the Recovery of Cs and Sr

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Paper 772 (extracted)

The purpose of this work was to design a simple selective separation procedure for ^{137}Cs and ^{90}Sr based on the use of mineral ion exchangers.[1-4]

The mineral ion exchangers suitable for work in an acid medium are generally the acid salts of tetravalent hydrolyzable metals, such as zirconium, titanium, and thorium (Ref. 5) and a series of heteropoly acids.[6-8] Of these materials, zirconium phosphate (ZrP)[5,9-13] displays the most interesting properties for ion-exchange work in acid solution; its high selectivity for cesium makes it particularly attractive. Very recently we discovered[14,15] that a new series of mineral ion exchangers, the ferrocyanide molybdates[16] (FeMo) and tungstates, exhibits still better characteristics with respect to cesium.

Owing to difficulties encountered with the separation of strontium in acid medium and in the presence of bulk ions, no fully satisfactory mineral ion exchanger has yet been found. The mineral clinoptilolite[25] shows some selectivity for this fission product in very dilute acid solution.

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Methods given for the preparation of ZrP yield diversified products (Refs. 9, 10, 12, and 17–20). The procedure used in this work gave a reproducible product independent of the type of zirconyl salt employed. The phosphorus-to-zirconium molar ratio in this product was 1.70 ± 0.1, and it had a total capacity of 4.5 ± 0.1 meq/g at *p*H 7. The chemical and ion-exchange properties are described in Refs. 12, 13, and 21 to 23.

The concentration of cesium in reprocessing effluents may vary from trace concentrations to 500 mg/liter, and the HNO_3 concentration, from 0.5*M* to 7*M*. Since ZrP shows no appreciable ion-exchange capacity above 2*M* HNO_3, we have restricted our investigation to 0.1*M* to 1*M* HNO_3. The results of these experiments show that the uptake of cesium increases with its concentration in solution at constant acidity and drops with increasing acidity.

The shape of the loading curves shows that dilution of a cesium solution does not improve the performance of the ZrP column. The maximum exchange capacities for a solution of 500 mg of cesium per liter are, respectively, 0.18, 0.24, 0.30, and 0.40 meq/g in 1*M*, 0.5*M*, 0.2*M*, and 0.1*M* HNO_3. These values represent the maximum attainable loading of cesium on ZrP and correspond to specific activities of 1.9, 2.5, 3.1, and 4.2 curies of ^{137}Cs per gram of dry ZrP.

Results are also given on the adsorption of cesium on ZrP from solutions having the same chemical constituents as reprocessing solutions from Hanford, Windscale, Marcoule, and Eurochemic.[24]

Mixing $NaMoO_4$ in HCl solution with $H_4Fe(CN)_6$ gives a precipitate of ferrocyanide molybdate (FeMo) which, after it has been filtered and dried, has an extremely high selectivity for cesium.[26]

The distribution coefficients for trace ions on FeMo show the product to have a very large total capacity and a fairly good available capacity for alkali and alkaline-earth metal cations in acid medium.[15] The most striking feature is the very high affinity of the exchanger for cesium. Results of a systematic study on the uptake from solutions containing 0 to 500 mg of cesium per liter showed that the exchange capacity is much higher than that with ZrP. From 10 to 21 curies of ^{137}Cs per gram of FeMo can be fixed in the presence of 2*M* to 0.1*M* HNO_3. Although these amounts drop in the presence of bulk ions, they still are five times higher than those for ZrP under similar conditions.

An experimental setup built for the separation of ^{137}Cs and ^{90}Sr with mineral ion exchangers is described.

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Recent Progress in Fission-Products Separation

By Andre Raggenbass*

Paper 100 (extracted)†

At the last Geneva Conference, we presented[1] a recovery method for ^{137}Cs based on the precipitation of cesium by phosphotungstic acid. Briefly, the cesium was precipitated as phosphotungstate, which was then decomposed by barium hydroxide. This yielded a mixture of cesium, barium (in excess), and ammonium hydroxides, the latter being expelled by distillation.

The barium ion was removed by precipitation with sulfuric or carbonic acid. The sulfuric acid method, tested in a pilot apparatus at Saclay, has given a regular supply of cesium sulfate; the carbonic acid method, giving cesium carbonate, has been tested on a semiindustrial scale. The barium carbonate can be precipitated in the presence of a precipitate of barium phosphate and tungstate; therefore all the operations can be performed in a single centrifuge with a minimum of transfers.

Many power reactors are designed to use alloys containing from 0.5 to 1% molybdenum. Such alloys are less degraded at high burnup levels than unalloyed uranium. However, the molybdenum interferes in the cesium-separation procedure outlined above. Complexing the molybdenum with iron was proposed at Geneva in 1958 (Ref. 2). By this process it was possible after concentration to achieve a final molybdenum content of about 20 g/liter, for a final iron concentration of 28 g/liter. Unfortunately later recovery of strontium was impossible in the presence of the large quantity of Fe(III) ions.

We now complex the molybdenum in the solution with phosphoric acid to form the phospho-12-molybdate ion. The solubility of this ion is such that the final solutions can contain 100 g of molybdenum per liter without risk of precipitation. Two radioactive ions are precipitated: cesium, which gives cesium phosphomolybdate, and zirconium, which gives zirconium phosphate. An alkaline treatment of this precipitate separates the cesium from the zirconium, which remains as a precipitate of zirconia. The total yield in the operation is 95%, and the ^{137}Cs contains less than 5×10^{-4}% of the original ^{144}Ce and less than 5×10^{-3}% of the original ^{90}Sr.

The exchange properties of the salts of heteropolyacids, such as phosphotungstates, were demonstrated several years ago;[3] unfortunately their physical nature prohibits their use in a fixed-bed exchanger. Attempts to incorporate a phosphotungstate or a phosphomolybdate in asbestos were unsuccessful. A parallel study with zirconium phosphate suggested that ammonium phosphotungstate would coprecipitate with zirconium phosphate. This is accomplished by precipitating the phosphate in a phosphotungstate suspension. Experiments have shown that this mixed exchanger has good mechanical properties, works well in a very acid medium, selectively adsorbs cesium, and has a capacity of about 0.1 meq per gram of exchanger.

The process we are investigating for the recovery of ^{90}Sr and the rare earths depends on the presence of large quantities of magnesium in our fission-product solutions. We have adopted D2EHPA as the extractant and have used it in variable proportions with the sodium salt according to the extractions desired. The diluent is *n*-dodecane to which is added TBP to avoid the appearance of a third phase. Flow sheets are given for each of the separation processes.

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†Editor's Note: The translation of Paper 100 received by the USAEC Division of Technical Information Extension did not contain the references. We have added those we could identify.

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Reprocessing of Uranium Fission-Product Solutions by Ion Exchangers

By O. Bobleter*

Paper 767 (extracted)

Ion exchangers, which are widely applied in numerous chemical separation and purification processes, have thus far been used for reprocessing uranium fission-product solutions only in certain areas and usually only for certain elements.[1-5]

Inexpensive organic substances were used as raw materials in several experiments for the production of exchangers to reduce the cost of waste processing. In some cases the regeneration of the exchanger should not be necessary. The raw materials included rotten wood,[6] lignite,[7] and peat.[8]

We used sawdust and lignin[9,10] to produce sulfonated cation exchangers. The best exchanger is obtained under the following conditions: 5 g of sawdust or lignin and 20 ml of a 10% sodium sulfite solution were adjusted with sodium hydroxide to *p*H 11 and were allowed to react in an autoclave for 3 hr at 150°C. Spruce sawdust and lignin gave exchange capacities of 0.43 and 0.67 meq/g, respectively. The production costs per milliequivalent of the exchanger were far below those of synthetic resin exchangers.

These products had good chemical stability and gave high decontamination factors, and incineration greatly reduced their volume. After preliminary experiments to determine the separation properties of fission products, a method of separating uranium fission-product solutions was developed that used only ion exchangers.

The separations were made in the following steps: (1) Uranium was separated from the bulk of the fission products on a Dowex 1-X8 anion exchanger. The column was eluted with a 4.2*M* $Al(NO_3)_3$ solution that left part of the Ce^{3+} on the column with the uranium.[11-15] (2) Uranium was separated from Ce^{3+} by adsorption on a sulfonated wood cation exchanger and elution with NH_4NO_3-HNO_3 solution.[16] (3) The ^{137}Cs, ^{90}Sr, and ^{90}Y were separated by adsorption on a sulfonated wood cation exchanger and elution with 0.015*N*, 0.14*N*, and 0.47*N* HNO_3, respectively.[17-19]

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Recovery and Purification of Fission Products from Irradiated Uranium

By Sten Ahrland and Karl Erik Holmberg*

Paper 609 (abstracted)

Methods of ion exchange with inorganic compounds and of freezing out of $UO_2(NO_3)_2$ from solution have been investigated as possible ways to process waste solutions and recover fission products.

Zirconium phosphate (ZrP), the hydrous oxides of zirconium, and silica gel are the inorganic ion exchangers of greatest promise.[1] Zirconium phosphate is moderately acidic, and the hydrous oxides are only weakly acidic. In fact, the zirconium hydrous oxide is amphoteric and acts mainly as an anion exchanger when *p*H is less than 5. The selectivity between ions of different charges generally increases with decreasing acidity. However, with more acidic gels a certain degree of sorption is reached at a lower *p*H value; this fact may be important for easily hydrolyzed ions. At a *p*H greater than 8, ZrP is heavily hydrolyzed and is thus not useful as an ion exchanger. Silica gel can be used up to a *p*H of 9, where solubility becomes appreciable. The workable ranges of *p*H and other properties of the ion exchangers investigated thus supplement each other rather well.

When a concentrated solution of $UO_2(NO_3)_2$ containing a small amount of fission products is partially frozen, $UO_2(NO_3)_2$ and ice crystallize, and the fission products are left in the mother liquid. The phases can be separated by centrifugation; the solid is left somewhat contaminated by the mother liquid. Repeated partial freezing should further purify the $UO_2(NO_3)_2$. The extent of separation of the more important fission products was tested, and decontamination factors of about 30 were obtained. The freezing out of $UO_2(NO_3)_2$ and ice may also be performed in the presence of moderate concentrations of HNO_3, which reduces the solubility of the $UO_2(NO_3)_2$.

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Separation of uranium from waste solutions by freezing has some advantages: the low temperature diminishes equipment corrosion, no chemicals or organic solvents are added, and freezing can be repeated many times for further purification. There are also some disadvantages: the decontamination factor is not very high although it might possibly be improved by using a higher centrifugal force or a longer centrifugation time, the freezing process is rather slow, and the remote handling of the solid can be difficult.

For the solid to be well separated from fission products, it must be melted and partially refrozen several times. It thus seems possible in principle to recover 95% or more of the $UO_2(NO_3)_2$ in a concentrated solution and to obtain it in a rather pure form by repeated freezings.

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Extraction of Some Elements with Phosphorus-Containing Monobasic Acids

By E. S. Gureev, S. M. Karpacheva, M. N. Ryjov, V. S. Smelov, E. G. Teterin, V. G. Timoshev, and I. K. Shvetsov*

Paper 347 (abstracted)

The extraction of a number of elements by phosphorus-containing acids is reviewed. In the first section the extraction mechanism of HNO_3, U^{6+}, Np^{5+}, Pu^{4+}, Cs, Am, Y, Ce, Pm, and Cm into this type of solvent is discussed. The second section summarizes the effect of the following factors on the extraction: organic-radical length, number of oxygen atoms (in going from phosphine oxide to phosphates), aromatic radicals, electronegative groups, radi-

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cal branching, and diluents. The references listed here are cited in the paper.

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Chemical and Electrochemical Behavior of Technetium in Aqueous Solutions

By Vict. I. Spitsyn, A. Th. Kuzina, N. N. Zamoshnikova, and A. A. Oblova*

Paper 349 (abstracted)

The concentration of technetium in the laboratory from molybdate and waste nitrate solutions is described. The methods discussed are based on extraction with acetone, quinoline, TBP, and isoamyl alcohol having extraction coefficients of 10^4, 10^3, 10^2, and 10, respectively.

The color reactions of technetium with furyldioxime, methyl violet, α-benzyldioxime, and orthocyclohexanedione-1,2-dioxime are the basis of the analytical methods for this element. The sensitivity of the reactions ranges from 0.7 to 0.1 μg/ml. Analytical methods for technetium that combine its extraction by acetone with spectrophotometry or beta counting have been developed.

Polarographic investigations were made of the electroreduction of pertechnetate, TcO_4^-, and hexachlorotechnetate, $TcCl_6^{2-}$, at the dropping-mercury electrode. The polarographic study of TcO_4^- was carried out in 1*M* solutions of KCl, KNO_3, and NH_4Cl over a range of *p*H values and in peroxide-alkaline and ammoniacal mediums.

Hexachlorotechnetate ions are formed in solution by treatment of pertechnetate with 4*M* hydrochloric acid at 90°C for 3 hr.

Tetravalent technetium as TcO_2 is oxidized by atmospheric oxygen in an alkaline medium to produce pertechnetate without the use of hydrogen peroxide.

The reduction of pertechnetate with hydrazine produces hexavalent technetium, and prolonged heating produces tetravalent technetium.

The electrodeposition of technetium in sulfurous acid or ammonium sulfate has shown technetium to be deposited on the cathode in a form which appears to be the oxide but which,

*Union of Soviet Socialist Republics.

on X-ray diffraction analysis, shows no crystalline structure. After the samples were heated in a vacuum, roentgenograms showed lines of both technetium metal and its dioxide.

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On the Behavior of Nitrosylruthenium Nitrato Complexes in TBP-HNO_3 System

By M. A. El-Guebeily, E. Hallaba, and Reda A. I. Azzam*

Paper 823 (abstracted)

The solvent-extraction behavior of the nitrato complexes of nitrosylruthenium in the TBP-HNO_3 system was studied. Both tracer ($10^{-9}M$) and macroscopic ($10^{-3}M$) concentrations of ruthenium were investigated. For tracer work the complexes were prepared by Martin's[1] method; for macroscopic work Fletcher's[2] procedure was used. A spectrophotometric study at $10^{-3}M$ ruthenium concentration of compounds prepared in both ways revealed that Fletcher's procedure gives directly the nitrato complexes and that Martin's method leads initially to RuO_4 in nitric acid which then undergoes a series of successive reductions leading to the nitrato complexes after 17 hr.

The extraction studies started with the TBP-H_2O-HNO_3 system without ruthenium and showed that the extractable species has the form $TBP(H_2O)_xHNO_3$, where x was found to be equal to 1 or 4, depending on the method of evaluation, with more evidence in favor of 4. The equilibrium constant k for the formation of this product was calculated. The effect of the following factors on the extraction behavior of nitrosylruthenium nitrato complexes at tracer and macroscopic concentrations was followed radiometrically: HNO_3 concentration, neutral nitrate-ion concentration, temperature, TBP concentration, and ruthenium concentration.

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PRODUCTION IN REACTORS

At the Second Geneva Conference in 1958, about 20 papers were presented on methods for the preparation of radioisotopes in reactors* and the synthesis of tagged compounds. At the Third Conference the number of papers on this subject was extremely small; most of the information was incidental to papers on research reactors. These papers show the increasing capability of various countries to produce their own supply of radioisotopes; summaries of pertinent parts are given here.

Objectives, Organizational Aspects, and Utilization of the IEA-R1 Research Reactor

By Azor C. Penteado Filho, Silvio B. Herdade, and Dirceu M. Vizeu†

Paper 118 (extracted)

The R1 research reactor at the Instituto de Energia Atômica (IEA) is a swimming pool type designed and built by the Babcock & Wilcox Co. for continuous operation at 5 Mw(t). The fuel elements are of the Materials Testing Reactor (MTR) type enriched to 20%. The reactor and its supporting facilities have been described previously.[1,2]

Regular production of ^{131}I at IEA began in 1960, and in 1961 ^{32}P was produced. Both isotopes were supplied mainly for medical applications.

The target materials for ^{32}P and ^{131}I are magnesium sulfate and telluric acid solution, respectively, since these offer certain advantages in chemical processing. About 200 g of magnesium sulfate and 300 g of telluric acid (the latter in aqueous solution having a concentration of 400 g/liter) are irradiated each week.

Radioactive gold and yttria seeds and millicurie quantities of ^{45}Ca, ^{60}Co, ^{76}As, ^{124}Sb, ^{131}Ba, ^{140}La, ^{170}Tm, etc., are also prepared on a routine basis for medical uses. Methods for producing colloidal ^{198}Au and high-specific-activity ^{51}Cr are also being developed.

Radiochemical processing takes place in a laboratory containing one cell reserved for the manufacture of ^{32}P and two for ^{131}I. The laboratory also houses a cell made ready for the production of ^{132}I from irradiation of uranium oxide.

The output of ^{32}P and ^{131}I for the past few years is as follows:

Isotope	Annual production, mc				
	1960	1961	1962	1963	1964*
^{32}P		670	1,140	1,180	300
^{131}I	164	2,500	14,400	23,300	6,400

*First 3 months only.

Compounds labeled with ^{131}I are the responsibility of the Radiopharmaceutical Section, which prepares labeled Rose Bengal, sodium orthoiodohippurate, human serum albumin, antipyrin, insulin, human growth hormone, polyvinylpyrrolidone, oleic acid, and triolein.

Materials for medical uses are checked for purity before dispatch, and those intended for injection are tested for sterility and absence of pyrogens.

Proc. U. N. Intern. Conf. Peaceful Uses At. Energy, 2nd Geneva, 20: 1-103, United Nations, New York, 1958.

†Instituto de Energia Atômica, São Paulo, Brazil.

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2. A. C. Penteado Filho and M. D. de Souza Santos, *Proceedings of the Symposium on Programming and Utilization of Research Reactors, Vienna, Oct. 16–21, 1961,* Vol. 2, pp. 397-408, Academic Press Inc., New York, 1962.

Scientific Activities in the Application of Atomic Energy for Peaceful Uses in Bulgaria

By G. Nadjakov*

Paper 879 (extracted)†

The Institute of Physics, Center of Atomic Research, Bulgarian Academy of Sciences, has established a laboratory for radiochemistry and radioisotope production. Its principal task is the production of necessary radioactive isotopes and the initiation of research in radiochemistry.

Owing to the activity of the laboratory in the last 2 years, the production of radioisotopes in the future is dependable in Bulgaria. The program of the laboratory has provided for the production of all short-lived isotopes and isotopes for medical purposes. This program, nevertheless, envisages the production of certain isotopes having long half-lives.

The laboratory produces the following isotopes for medical use: ^{24}Na (as chloride tablets), ^{42}K (as chloride tablets), ^{90}Y (as oxide), and ^{198}Au (as pellets and in colloidal solution).

The following radioisotopes destined to be used as tracers or for other purposes are produced in limited quantities:

1. Sources of pure beta radiations: Phosphorus-32 (as sodium or potassium salts), ^{35}S (carrier free, as sulfuric acid and as barium sulfate), and ^{45}Ca (as chloride or nitrate solution or calcium carbonate).

2. Sources of gamma radiations: ^{82}Br, ^{187}W, ^{72}Ga, ^{131}I, ^{192}Ir, ^{60}Co, ^{140}La, ^{56}Mn, ^{64}Cu, ^{86}Rb, ^{46}Sc, ^{182}Ta, ^{141}Ce, and ^{65}Zn.

The average annual shipment of different isotopes is about 30 curies of isotopes for medical purposes, 80 curies of neutron sources of polonium-beryllium and radium-beryllium for geological prospecting, and 6000 mg radium equivalent of ^{60}Co. In addition to these isotopes, approximately 250 standards of alpha-beta preparations have been supplied. Also approximately 40 different isotopes are imported into the country annually. In 1964 the laboratory of radiochemistry attached to the IRT-1000 reactor at Sofia produced about 20 different short half-lived isotopes of several hours to 3 days, such as ^{198}Au, ^{24}Na, and ^{42}K. These isotopes will meet all the needs of hospitals and research institutes.

*Director of the Institute of Physics, Center of Atomic Research, Bulgarian Academy of Sciences; Vice President of the Committee for Utilization of Atomic Energy for Peaceful Uses, Council of Ministers, People's Republic of Bulgaria.

†Report ORNL-tr-418, originally in French. Translated in part by D. A. Fuccillo, Jr., ORNL, Isotopes Information Center, Nov. 23, 1964. This paper also contains a lengthy list of all the medical, biological, and agricultural research projects using radioisotopes now under way in Bulgaria.

Research Reactor and Its Utilization in the Republic of China

By Chen-hwa Cheng*

Paper 481 (extracted)

The type and size of the Chinese research reactor were decided by considering its flexibility in meeting different purposes (such as training, research, and isotope production), its safety, and its cost. A 1-Mw light-water-moderated and -cooled graphite-reflected open-pool heterogeneous reactor was chosen after careful study and discussion with USAEC advisers. Through open bid a contract for designing and supplying the reactor components was awarded to the General Electric Company. However, Tsing Hua University assumed the responsibility for engineering design and erection work as well as for the design and execution of the reactor housing.

The use of short-lived isotopes as tracers in chemical experiments may not be feasible unless the experiments are conducted close to a

*Director, Institute of Nuclear Science, National Tsing Hua University, Hsinchu, Taiwan, China.

reactor center. If a reactor is available, then short-lived isotopes are often preferred because they can be prepared in high specific activity by short irradiations. Hence production of short-lived radioisotopes is one of the important applications of the reactor.

From a survey conducted by the Institute on the uses of radioisotopes in Taiwan, it was decided to produce the following species of radioisotopes: ^{32}P, ^{35}S, ^{131}I, ^{131}I-Rose Bengal, colloidal ^{198}Au, ^{24}Na, ^{42}K, ^{59}Fe, ^{51}Cr, ^{203}Hg, ^{45}Ca, and ^{86}Rb. Radioisotopes produced and supplied to various institutions and hospitals during the period Apr. 24, 1962, to Apr. 15, 1964, are given below:

Radio-isotope	Amount, mc	Destination
^{131}I	4330	National Taiwan University Hospital National Taiwan University Medical School Taiwan Sugar Experimental Station National Taiwan University, School of Agriculture
^{198}Au (colloid)	1500	Veterans Hospital Taipei Provincial Hospital National Taiwan University Hospital
^{24}Na	280	U. S. Navy Medical Research Unit, No. 2
^{42}K	150	U. S. Navy Medical Research Unit, No. 2 Taiwan Sugar Experimental Station
^{32}P	118	National Taiwan University, School of Agriculture Taiwan Sugar Experimental Station National Taiwan University, School of Chemistry
^{35}S	20	National Taiwan University, School of Agriculture
^{51}Cr	30	National Taiwan University Hospital
^{59}Fe	10	National Taiwan University Hospital

Five Years' Experience with the Hungarian WWRS Type Reactor

By F. Szabo and L. Varkonyi*

Paper 451 (extracted)

The WWRS type research reactor in Budapest, which went critical in March 1959, is operated by the Central Institute of Physical Research. The reactor is used not only by the research laboratories of the Institute but also by the Isotope Institute, the Frederic Joliot-Curie Research Institute for Radiation Biology and Health, and other institutes and industrial works.

Preparations for physical research and isotope production started almost simultaneously with the building of the reactor. When the operation schedule was being devised, one consideration was that this reactor, being the only one in the country, ought to be available both for research and for isotope production. The use of the reactor has grown and now seems to be nearing its limit.

Isotope production has been one of the primary functions of the reactor since 1960; the production capacity of the reactor, however, does not cover the entire demand of the country. Careful considerations have prompted the following programming priority:

1. Production of short-lived isotopes, the transport of which is costly, complicated, and sometimes even impossible
2. Production of high-purity or high-specific-activity isotopes to meet special demands.

Relevant data on isotope production are given in Table 451-1.

*Central Institute of Physical Research, Budapest, Hungary.

Six Years of Utilization of the Polish Research Reactor EWA

By J. Aleksandrowicz, B. Buras, R. Plejewski, and S. Siekierski†

Paper 119 (extracted)

The Polish research reactor EWA at Swierk is a light-water-moderated and -cooled 10% enriched-uranium 2-Mw tank type reactor with an average thermal-neutron flux of 10^{13} n $(cm^2)(sec)$. The reactor, received from the USSR, became critical in 1958 and since then has worked smoothly at full power.

Radioisotope production in the EWA started first with sealed sources and now almost

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Table 451-1 RADIOISOTOPES PRODUCED IN HUNGARY

Isotope	Production 1960 Curies	1960 mc	1960 μc	1961 Curies	1961 mc	1961 μc	1962 Curies	1962 mc	1962 μc	1963 Curies	1963 mc	1963 μc
^{110}Ag		756	200		500	560		386	500		158	780
^{111}Ag		36	200		97			188	600		244	900
^{198}Au	13	351		12	692	500	9	694	60	9	758	510
^{140}Ba		124			227	740	1	965	690		556	250
^{51}Cr	1	309	780	2	82		2	614	960	2	115	630
^{64}Cu		113	800		193	719		23	340		244	135
^{197}Hg											337	890
^{203}Hg		66			66	350		77	190		186	770
^{166}Ho					15	600		250	530		2	
^{131}I	12	80	450	10	529	430	27	357	950	42	622	326
^{192}Ir					381			353			10	
^{42}K	1	217	200	2	553	750	1	585	950	1	318	860
^{140}La			500		285			100			25	600
^{177}Lu					10	300		305	100		1	
^{24}Na	1	193		3	718	130	3	440	350	1	970	720
^{32}P											610	
^{86}Rb								206			805	280
^{161}Tb											481	350
^{170}Tm					108	200		884	840		887	300
^{90}Y	1	19	550	1	287	700		782	40	1	665	550
^{65}Zn											791	94
Total*	31	267	680	34	748	979	50	216	100	64	793	945

*Includes small amounts of the following isotopes whose annual production is less than 100 mc: ^{76}As, ^{115}Cd, ^{169}Er, ^{152}Eu–^{154}Eu, ^{114}In, ^{99}Mo–^{99}Tc, ^{147}Nd, ^{147}Nd–^{147}Pm, ^{210}Po, ^{124}Sb, ^{75}Se, ^{85}Sr–^{89}Sr, ^{175}Yb, and ^{175}Yb–^{169}Yb.

completely meets the demand of the country. Sources have been used in all kinds of gamma-ray relays and in instruments for various measurements, such as thickness, density, and liquid levels. The main sealed sources produced are ^{60}Co, ^{75}Se, ^{110}Ag, ^{65}Zn, ^{170}Tm, and ^{192}Ir. From 1500 to 2000 sealed sources are produced each year.

Cobalt-60 sources for radiography are not produced; however, the specific activity of the ^{192}Ir obtained is high enough for radiography (e.g., a cylindrical source 2 mm in diameter and 2 mm long has an activity of 5.5 curies).

Apart from typical sealed sources, the reactor has been used also for irradiation of a wide range of target materials to be used as tracers. The number of targets irradiated in the reactor is steadily increasing, and in 1963 it amounted to about 500. The range of the source activity obtained varied from 0.1 mc to 10 curies. The target materials contained nearly all kinds of radioisotopes with half-lives greater than several hours. Bromine-82, for example, is used in amounts of several curies in hydrological investigation. Sodium-24 and ^{65}Zn are used as tracers for estimating the material-transport rate in technological processes. Also machine parts are irradiated for wear tests.

An important part in the production of radioactive materials is the preparation of carrier-free solutions. Iodine-131, obtained by irradiation of TeO_2 and wet extraction of the ^{131}I, is produced in amounts of 1 to 2 curies/week, mainly for medical purposes. The iodide is also used for production of ^{131}I-labeled organic compounds, such as albumin, Rose Bengal, oleic acid, diiodotyrosine, and Hippuran. From 150 to 200 mc of these compounds is produced per month.

Phosphorus-32 (about 200 to 300 mc/week) and ^{35}S (about 0.5 curie/month) are produced for medical and agricultural purposes. Solutions of ^{42}K, ^{24}Na, ^{51}Cr, ^{65}Zn, ^{45}Ca, ^{198}Au, and ^{64}Cu are systematically produced. The activity is from several millicuries to several hundred millicuries per month.

Occasionally other nuclides are also produced from time to time on special orders of about 100 mc. In this group are nuclides obtained in the (n,γ) and (n,p) reactions (e.g., ^{58}Co, ^{54}Mn, and ^{59}Fe). During the 6 years of reactor operation, targets of aluminum and lithium-aluminum alloy have been continually irradiated; hence the extraction of ^{14}C (*sic*) and ^{3}H will be possible in the near future. The estimated activity of ^{14}C will be 2 to 2.5 curies, and that of tritium, several hundred curies.

Research with the VVR-S, Experimental Reactor of the Atomic Physics Institute in Bucharest

By Horia Hulubei*

Paper 189 (extracted)

The Romanian VVR-S is a 2000-kw(t) 10% enriched-uranium ordinary-water-moderated and -cooled experimental reactor. It was supplied by the USSR and is the first concrete step toward realization of a project submitted by the Soviets to the 1955 Geneva Conference.

The VVR-S became critical in July 1957, and radioisotope production began as soon as the reactor was put into operation. Emphasis was given to this activity because of economic considerations and because of the large potential application of nuclear physics methods to scientific research and to industry. Work with radioisotopes is a practical means for training specialists in nuclear physics and for arousing public interest.

The demand for radioisotopes has increased each year, and at present almost 50 different radioisotopes are regularly supplied by the Institute. By the end of the year, it is anticipated that 600 curies will be supplied in various forms.

At present no fission products are produced, but plans are to turn over to a specialized industry the production and supply of radioisotopes.

Radioactive substances that are not produced within the country for economical reasons are imported.

Research Reactors in Spain

By C. Sánchez del Río, J. Montes, F. de la Cruz, G. Dominquez, J. C. de Zabalo, and J. J. Clúa†

Paper 492 (extracted)

At present Spain has three research reactors, JEN-1, ARGOS, and ARBI. The JEN-1, located in the National Center of Atomic Energy in Madrid, was the first reactor installed; it is a 3-Mw swimming pool type and was put into operation in the fall of 1958.

So that the JEN-1 could be used for the production of radioisotopes, a laboratory for handling up to 100 curies of radioactive materials together with the corresponding shielded areas and control equipment was designed and built. Production methods have been studied for those isotopes which can best be produced with this reactor.

Table 492-1 PRODUCTION AND CONSUMPTION OF RADIOISOTOPES IN SPAIN DURING 1963

Isotope	Imported, mc	JEN-1 produced, mc	JEN-1 produced, % of total
^{14}C	21		
^{32}P	602	1.134	65
^{35}S	1	45	98
^{45}Ca	0.5		
^{51}Cr	15	16	51
^{54}Mn		0.05	100
^{58}Co	0.02		
^{59}Fe	0.2	0.44	69
^{60}Co	10.714 curies		
^{85}Kr	450		
^{90}Sr	996		
^{124}Sb		6.0	100
^{131}I	3.835	6.277	62
^{137}Cs	15.110		
^{144}Ce	44		
^{192}Ir	150.4 curies	49	0.3
^{198}Au	2.360	8.878	79
^{203}Hg	102	20	16
^{204}Tl	63		
^{226}Ra	417		
^{241}Am	30		

The selection of materials to be produced was made after a careful consideration of the isotope-consumption statistics in Spain and abroad and after a detailed study of the irradiation possibilities and anticipated operation rate of the JEN-1. The isotopes selected were ^{32}P, ^{35}S, ^{51}Cr, ^{131}I, and ^{198}Au (both colloidal and in pellets). Occasionally other isotopes and compounds labeled with ^{14}C, ^{131}I, ^{32}P, ^{35}S, and ^{15}N are produced. Such isotopes as ^{60}Co and ^{192}Ir with high specific activity cannot be produced in JEN-1. Equipment is being built for the preparation of ^{14}C as $BaCO_3$ and of ^{3}H as gas or water.

Data on the Spanish isotope market in 1963 are given in Table 492-1.

*Atomic Physics Institute, Bucharest, Romania.
†Junta de Energía Nuclear, Madrid; Escuela Técnica Superior de Ingenieros Industriales, Bilbao; Escuela Técnica Superior de Ingenieros Industriales, Barcelona, Spain.

Erection, Testing, and Operation of the Radioisotope Production Facility of the UARAEE

By M. A. El-Guebeily, E. Hallaba, and M. El Garhy*

Paper 816 (extracted)

A 2-Mw(t) VVRS type reactor was put into operation by the United Arab Republic Atomic Energy Establishment (UARAEE) in July 1961 in cooperation with the Soviet Atomic Energy Authority. A detailed description of this reactor is given in several papers.[1,2] Among its various facilities this reactor has eight vertical channels for the production of radioisotopes and for various investigations in the field of nuclear chemistry. The neutron flux is 2×10^{13} n/(cm^2)(sec) in the core.

The initial program started with the production of the most commonly used short-lived processed isotopes and has been implemented by a development program covering production of a number of other processed and unprocessed isotopes and radiation sources. A description of this project is presented in Ref. 3. Construction of the isotope-production facility started in July 1961 in cooperation with IFA and the Norwegian firm NORATOM A/S, and the laboratory was commissioned in December 1962.

The radioisotope laboratory includes facilities for the routine production of processed and unprocessed radioisotopes. The production capacity for the main processed isotopes is as follows:

Isotope	Half-life	Number of cells	Capacity/cell/batch, curies
^{42}K	12.5 hr	1	2
^{24}Na	15 hr	1	0.05
^{198}Au (colloid)	2.7 days	1	5
^{198}Au (Muller)	2.7 days	1	5
^{131}I	8 days	2	1
^{32}P	14.2 days	1	0.5
^{51}Cr	27.8 days	1	0.6
^{35}S	87 days	1	0.5

*Nuclear Chemistry Department, United Arab Republic Atomic Energy Establishment, Inshas, Egypt.

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1. I. Hammouda et al., this Conference, Paper 453.
2. V. V. Goncharov et al., *Proc. U. N. Intern. Conf. Peaceful Uses At. Energy, 2nd Geneva*, 10: 321-67, United Nations, New York, 1958.
3. H. Mahmoud et al., *Proceedings of the Symposium on Programming and Utilization of Research Reactors, Vienna, Oct. 16–21, 1961*, Vol. 3, pp. 395-403, Academic Press Inc., New York, 1962.

Programs for Utilization of the Research Reactor TR-1 at the Cekmece Nuclear Research Center

By Sait Akpinart

Paper 747 (extracted)

The Çekmece Nuclear Research Center, whose laboratory buildings are still under construction, was inaugurated on May 27, 1962. Presently it is the only institution operated directly by the Turkish Atomic Energy Commission (TAEC). The principal facility of the Center is the Turkish Reactor No. 1 (TR-1).

The TR-1 is a heterogeneous light-water-moderated and -cooled 1-Mw swimming pool reactor with enriched uranium as fuel. It was designed and constructed during 1959–1961 by AMF Atomics, Inc., with local subcontractors on a turn-key basis. The startup experiment was performed on Jan. 6, 1962.

The Chemistry Department uses the reactor for gamma irradiations and radioisotope production. Up to now most radioisotope requests from hospitals and university laboratories have been met since the volume of the demand has not been high and, in general, the activities required have been low. However, a country-wide survey indicates that the demand will increase with time to justify a special laboratory for isotope production. A temporary, inexpensive prefabricated building was erected close to the reactor to handle requests more effectively and safely. The low-activity chemistry laboratories are expected to be completed by the end of this year. The chemistry group has received help from Brookhaven National Lab-

†Çekmece Nuclear Research Center, Istanbul, Turkey.

oratory in neutron-irradiation techniques and valuable advice from IAEA experts in problems connected with radioisotope production.

Isotope distribution in 1963 is given below:

Isotope*	Activity, mc	User
^{198}Au	4	Ege University, Izmir
	15	Technical University, Istanbul
^{198}Au seeds	2	Gureba Hospital, Istanbul
^{192}Ir wire	100	Gureba Hospital, Istanbul
^{32}P	22	Ege University, Izmir
	15	Technical University, Istanbul
^{65}Zn	1	University of Istanbul, Istanbul
	3	Ege University, Izmir
^{45}Ca	120	Ege University, Izmir
^{51}Cr	10	Ege University, Izmir
	2	Robert College, Istanbul
^{24}Na	730	Topkapi Palace Museum

*Isotopes produced for different groups at the Center and irradiations made for the Center and for various universities are excluded from the table.

RESEARCH REACTORS

Research reactors are used in many countries for the production of radioisotopes. Descriptions of these reactors are given in the papers listed here. The list includes papers on well-established reactors that are used in the routine production of isotopes and on new experimental very high flux reactors that may be important in the production of isotopes in the future.

1. K. N. Beckurts, N. Fiebiger, E. Fischer, L. Koester, T. Springer, K. Wirtz, and K. E. Zimen, Research Reactors in the Federal Republic of Germany, Paper 534, Germany.
2. M. Bogaardt, J. Coehoorn, W. Hofman, and W. W. Nijs, The Biological Agricultural Reactor Netherlands (Barn) Design and Commissioning, Paper 725, Netherlands.
3. C. de Chauvez and F. Rossillon, Use and Evolution of the Research Reactors of the C. E. A., Paper 33, France.
4. D. R. deBoisblanc, The NRTS Advanced Test Reactor, Paper 223, USA.
5. V. V. Goncharov, Review of Research Reactor Activities in the Soviet Union, Paper 296, USSR.
6. J. M. Hendrie, The Brookhaven High Flux Beam Research Reactor, Paper 222, USA (also USAEC Report BNL-7960).
7. D. G. Hurst, R. E. Manson, E. Critoph, and H. E. Petch, Utilization of Canadian Research Reactors, Paper 2, Canada (also Canadian Report AECL-2002).
8. R. J. Jackson (Ed.), Experience of Operation and Use of the U. K. High-Flux Research Reactors, Paper 123, England.
9. H. Kouts, The Uses and Potentialities of Research Reactors, Paper 194, USA.
10. A. K. Krasin, V. A. Naumov, I. A. Savushkiv, G. I. Strelkov, and O. I. Yaroshevich, Physical Characteristics of Research Pool-Type Reactor IRT-2000 with Test-Loop Channel, Paper 718, USSR.
11. F. Musilek, F. Berger, L. David, Z. Hrdlicka, B. Stoces, J. Urbanec, and V. Zajic, A Review of Investigations and Experimental Utilization of the Research Reactor in the Institute of Nuclear Research of the Czechoslovak Academy of Sciences, Paper 531, Czechoslovakia.
12. A. G. Ramalho, Portuguese Research Reactor, Paper 505, Portugal.
13. R. Ramanna, Utilization of Research Reactors at Trombay, Paper 744, India.
14. Research Reactors in Japan, Paper 120, Japan.
15. F. Rossillon and C. de Chauvez, A Survey of Experimental Facilities and Flux Measurement Used in C. E. A. Research Reactors, Paper 34, France.
16. J. J. M. Snepvangers, M. Grenon, H. Postma, and K. F. Maartman-Moe, Utilization of the High Flux Reactor at Petten, Paper 724, Netherlands.
17. J. A. Swartout, A. L. Boch, T. E. Cole, R. D. Cheverton, G. M. Adamson, and C. E. Winters, The Oak Ridge High Flux Isotope Reactors, Paper 221, USA.

ISOTOPE SEPARATION

Several papers on methods for the separation of stable isotopes were given. These papers are listed with a brief note of their contents to help those interested in this field.

General Papers

1. F. Botter, P. Molinari, and G. Dirian, Physico-Chemical Separations of Stable Isotopes, Paper 90, France. Separation of hydrogen isotopes by thermal diffusion and chromatography. Separation of lithium isotopes by ion exchange and preparation of ^{46}Ca by countercurrent migration of fused salts.
2. A. O. Edmunds, E. Blancke, and E. Rogers, The Economic Separation of Stable Isotopes on a Commercial Scale, Paper 567, England. Separation of boron isotopes by distillation of BF_3 and $(CH_3)_2O \cdot BF_3$. Separation of ^{13}C by distillation of CO.
3. I. G. Gverdtziteli, T. A. Gagooa, G. N. Muskhelishvili, U. V. Nikolaev, E. D. Oziashvili, and G. A. Thevzadze, Experimental Production of Stable Isotopes, Paper 388, USSR. Separation of boron isotopes by distillation of $(CH_3)_2O \cdot BF_3$. Nitrogen-15 preparation by chemical exchange. A brief discussion on separation of isotopes of neon, carbon, argon, and krypton by mass diffusion.
4. M. Silvestri, G. C. Angelino, and B. Brigoli, A New Distillation Technique for Isotope Concentration, Paper 557, Italy. Separation of oxygen isotopes by distillation of water.

5. M. M. Urgell, J. Iglesias, J. Casas, J. M. Savirón, and M. Quintanilla, The Production of Stable Isotopes in Spain, Paper 491, Spain. Separation of nitrogen and boron isotopes by ion exchange and of neon isotopes by thermal diffusion.

Gas Centrifuge

6. J. Jordan, W. Kolbe, and R. Borges da Silva, Separation of Stable Isotopes by the Gas Centrifuge Method. Influence of the Axial Pressure on the Separation of Argon Isotopes, Paper 482, Brazil. Separation of argon isotopes by Groth type centrifuge.
7. A. Kanagawa, M. E. Yoichi Takashima, and Y. Oyama, Isotopes Separation by Concurrent Gas Centrifuge, Paper 440, Japan. Mostly theory; small amount of data on separation of uranium isotopes.
8. C. Ouwerkerk and J. Los, The Separative Power of Short Countercurrent Centrifuges, Paper 637, Netherlands. Entirely theoretical.

Lithium Isotopes

9. G. J. Arkenbout, Separation by Countercurrent-Exchange-Electrolysis Applied on Lithium Isotopes, Paper 639, Netherlands. Electrolysis and countercurrent exchange between lithium amalgam and LiBr solution.

Uranium Isotopes

10. C. Frejacques and R. Galley, Deductions Based on Studies of Uranium Isotope Separation and French Achievements in This Field, Paper 89, France. Mainly theory and economic data on gaseous diffusion. A small amount of information on separation of uranium by gas centrifuge.
11. H. Kakihana, Some Japanese Works on Chemical Separation of Uranium Isotopes, Paper 439, Japan. Use of ion exchangers and chemical exchange to attempt the separation of uranium isotopes.

Deuterium Separation

12. J. Alvarez, M. Gispert, A. Maria, J. L. Otero, and J. L. Rojas, Studies on the Production of Heavy Water at J. E. N., Paper 496, Spain. Separation by electrolysis on a pilot-plant scale.
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Section

III

Isotopes and Radiation Technology

Isotope Technology Development

In this section papers from three different phases of isotope technology are presented: hydrology, engineering applications, and concentration or recovery of isotopes for gas counting. Where the report has been shortened, condensed, or abstracted, methodology has been retained but detailed results omitted. A review of the role of radioisotope techniques in hydrology is presented in one paper. One of the two reports concerning the recovery of radioisotopes for gas (proportional) counting is pointed toward hydrological applications; however, the technique may also be appropriate in other fields. The engineering papers are concerned with the use of isotopes in road construction, the determination of pipeline wear in iron-ore transportation, the measurement of gamma doses by the conductivity of cuprous oxide, and the choice of radiation characteristics for various gaging problems.

The Role of Radioisotope Techniques in Hydrology

By B. R. Payne, J. F. Cameron, A. E. Peckham, and L. L. Thatcher*

Paper 875

There is no doubt that water is the prime requisite for development of any area, but it is only recently that widespread measures have been taken to ensure its most efficient use. In 1965 the International Hydrological Decade will begin under the auspices of UNESCO. The main objective will be to accelerate the study of the water resources with a view to their rational development and use. When the program was drawn up, it was recognized that more emphasis must be placed in certain areas, such as groundwater. During the Decade isotope techniques will be widely used. This paper reviews the role that these techniques are already playing and emphasizes how scientific hydrology can benefit by a wider use of these relatively new methods.

Application of Radioisotopes in Groundwater Hydrology

The use of groundwater is based on a knowledge of its origin, direction and velocity, quantity, accessibility, and quality. Radioisotopes, because of their high sensitivity of detection, are now being used for the measurement of these parameters. The applications may be considered under three headings: (1) localized groundwater studies involving the use of artificially injected radioisotopes; (2) regional groundwater studies where use is made of environmental radioactivity; and (3) borehole logging in measuring lithological characteristics relevant to groundwater investigations.

An important measurement frequently facing the groundwater hydrologist is the velocity of movement of water through an aquifer. This may be regional movement from a large recharge area, which gives information on the rate at which an underground reservoir is replenished, or it may be local movement at a single well, which gives information on the yield of that well. Conventional methods of estimating velocity of water movement include computations from head and permeability considerations, pumping tests, and dye or salt-tracer movement between wells. Pumping tests are costly and not always applicable. Dye and salt tracing suffer from retardation effects,

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density effects, and poor detection sensitivity. Radioisotope techniques for velocity measurement offer advantages in both the localized and regional applications. Hence this is one of the major applications of radioisotopes in groundwater hydrology.

LOCALIZED GROUNDWATER STUDIES

Single-Well Method for the Measurement of Groundwater Velocity. In 1957 an isotopic method for determining the rate of horizontal groundwater flow was developed by Raymond and Bierschenk[1] and by Moser et al.[2] This is the single-well method, which involves the injection of a radioisotope solution into a confined section of a well and the measurement of the exponential dilution of the isotope solution with time as unlabeled water slowly moves into the well. The expression for the calculation of velocity is

$$v_g = -\frac{V}{\psi F t} \times \ln \frac{C}{C_0}$$

where v_g = the groundwater velocity
V = the dilution volume
F = the cross-section of volume V
t = time
C = tracer concentration after t
C_0 = tracer concentration for $t = 0$
ψ = coefficient taking into account the deformation of the hydrodynamical field due to the presence of the hole. The coefficient ψ is defined by the equation $Q = \psi Q_w$, where Q is the flow rate of water passing through the cross-section of the well and Q_w is the flow rate of water passing through the same cross-section of the aquifer.

The derivation of this equation assumes flow through a section of a cylinder at right angles to the axis. Therefore it applies to horizontal flow in wells, and successful application to field measurements requires that vertical currents be insignificant. This is achieved in practice by confining a well section with seals at the top and bottom of the velocity probe.

The evaluation of the coefficient ψ under widely varying underground conditions remains the single most difficult aspect in the application of the point-dilution method. Research that may extend the method to new problems is sponsored by the International Atomic Energy Agency[3] and is directed toward the analysis of the parameters that affect ψ. Ogilvi[4] determined from theoretical considerations values of ψ for a wide range of conditions. Even the uncorrected measured velocity, which has been termed "filtration velocity," is very useful since it provides yield information that can significantly reduce costly pumping tests.

Model tanks have been used at the Bundesversuchs- und Forschungsanstalt, Vienna,[5] to test the application of Ogilvi's equation to sand and gravel aquifers found in Austria. The equation was generally confirmed, and it was found in practice that filter tubes with 8 to 10% perforation were satisfactory at velocities up to 0.1 cm/sec. At this point 30% perforation becomes necessary. At velocities below 10^{-4} cm/sec, correction for diffusion was made.

Development of refined point-velocity probes has continued since the introduction of the first instruments in 1957. A schematic representation of Mairhofer's probe[5] is shown in Fig. 875-1. This unit eliminates the mixing problem by use of a premixed isotope solution that displaces almost all the volume in the section

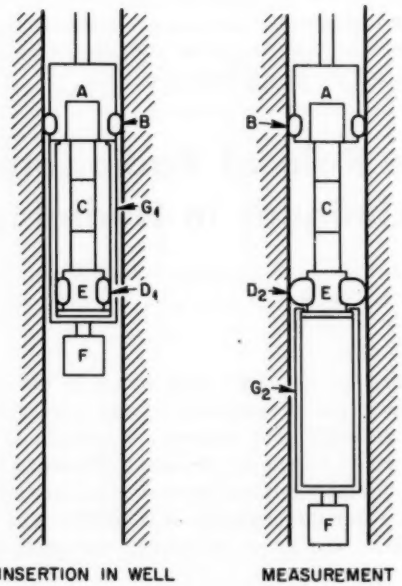

Fig. 875-1 Mairhofer's design of a point-dilution velocity probe. A, electronics head; B, upper rubber seal; C, detector; D_1, lower rubber seal, deflated; D_2, lower seal, inflated; E, electromagnet release; F, weight; G_1, outer housing, insertion position; G_2, outer housing, dropped.

to be tested. The solution is exposed to the groundwater stream by dropping the outer shell. Guizerix et al.[6] used an electrically driven stirring vane in the probe to maintain continuous mixing throughout the measurement. Moser[7] has developed vertical velocity probes and an injector that permits sequential injections of isotope solution without removing the unit from the well.

The isotopes most frequently used are ^{131}I and ^{82}Br. Tritium has been used in a technique that involves the continuous removal of samples from a well, and ^{86}Rb has been used in a situation where ^{131}I is highly adsorbed. Using adsorption properties of iodine constructively, Mairhofer pioneered the measurement of the direction of groundwater flow. Radioisotope cations, such as silver, are also used. The isotope is injected into the well section under study and then allowed to drift out and become adsorbed in the exposed aquifer. The section is scanned radially with a collimated detector, and the direction of exit of the isotope is determined.

In the Federal Republic of Germany,[8] the single-well method has been used to establish seepage patterns at the Rosshaupten Dam at a depth of 40 m below the storage level. Similar tests were made at the Sylvenstein Reservoir,[9] and at another dam it was demonstrated that remedial measures had blocked under-seepage paths but new seepage had developed laterally around the structure. These tests involved the measurement of vertical flow and were successfully accomplished at a depth of 250 m. In Vorarlberg, Austria, results by the vane-meter and the single-well methods gave good agreement. These tests involved permeability measurements in which water is pumped to create a vertical flow. Mairhofer has applied the single-well method to determine the relative yields of wells supplying Vienna.

In Poland, Borowczyk et al.[10] have continued the development of the single-well method and have analyzed the effect of the detector-tube volume on flow equations. Królikowski[11,12] has made further studies of the single-well technique.

Several specialized variations of the versatile single-well technique have been developed for specific problems.

Radioisotopes are being used in Israel in a U. N. Special Fund project to investigate problems of changes in the salinity of groundwater induced by artificial recharge of aquifers. Suitable techniques are being developed for control of quality of groundwater by a carefully planned system of pumping. Where the chloride content of the waters cannot be used as a tracer, ^{60}Co as potassium cobalticyanide is used to label large volumes of water that are being used for recharging through a pumping well. The experimental data are being used in theoretical and model studies and are also being considered as a means of control of the final operational program.

A significant combination of radioisotope techniques with standard pumping tests has been reported by Degot et al.[13] They used ^{82}Br in pumping tests to obtain directional information and to find the maximum velocity in two superimposed aquifers in addition to the average velocity normally obtained in a pumping test. Directional flow measurements were also made with ^{82}Br in three injection wells around the pumping well. The system was analyzed in less time and in more detail than would have been possible by the pumping test alone. Degot et al.[13] have also reported the use of ^{82}Br to plot in detail sharp changes in the horizontal permeability with depth in a series of boreholes in the harbor sediments off Boulogne. The relative variations of permeability with depth were clearly disclosed in detail.

A second method for the detailed examination of stratified water flow in wells has been presented by Bird and Dempsey,[14] who used the method to identify permeable strata for water flooding as a means of secondary recovery operations in oil fields. Minute spherules containing a radioisotope were used. As the water under pressure moves into the permeable layers, the spherules are filtered out on the face of the exposed aquifer in proportion to the permeability. The hole is then scanned with a collimated detector. Figure 875-2 shows the kind of plot obtained.

Groundwater Tracing. A U. N. Special Fund project in Greece[15] was intended to explore and develop the water resources of karstic terrain in the Peloponnese. Tritium tracing was used to investigate connections between sinkholes on the high plateau of Tripolis and springs, some of which issued under the sea. Two injections of tritiated water totaling 1400 curies gave the following significant results:

1. Tritium could be successfully used to trace underground water movement over a

distance of 30 km in karstic terrain where fluorescein gave no result.

2. Dual tracer detection peaks at one spring revealed two distinct underground channels.

3. From the accumulated water discharge and the shape of the tritium discharge curve, a stored volume of 30,000 m^3 between a major sinkhole and spring was established.

4. The discharge of an underwater spring that could not be measured by conventional methods was successfully measured.

Lapointe[16] reports the use of tritium tracer to investigate leakage into a mine. A mine in Ontario province was troubled by seepage. A lake 200 m away, the suspected source, was labeled with tritium, and groundwater samples were taken between the lake and the mine. No tritium increase was observed; therefore the lake was ruled out as a source. Tritium added to a sump in the rock wall outside the ore body revealed the true seepage path.

A method for the measurement of groundwater velocity in different strata has been advanced by Iwai and Inoue,[17] who have tested it in a model tank with two sand layers of different packing that were separated by an impermeable membrane. A mixture of tritiated water and ^{90}Sr was introduced into the system, and the respective breakthrough curves were observed. Substitution into an equation that includes factors for the relative adsorption holdup of the two tracers gives the velocity stratification. The method appears to involve several practical limitations that will have to be resolved for field application.

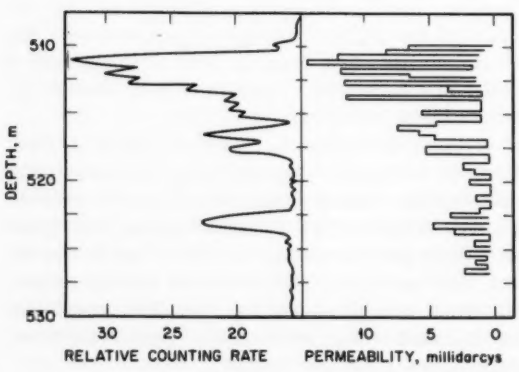

Fig. 875-2 Stratification of permeability (particulate tracer).

REGIONAL GROUNDWATER STUDIES

In regional hydrologic methods the tritium technique introduced by Libby is unquestionably the most significant. Developments in recent years, however, have demonstrated the necessity for the closest cooperation between the hydrologist and the isotope specialist for the intelligent application and interpretation of this specialized tool. As an example, Theis has called attention to the great significance of dispersion effects that spread the tracer front both laterally and longitudinally as it moves down gradient. Theis[18] provided a new interpretation of the results obtained by von Buttlar[19] in his studies of tritium in groundwater of the Carrizozo area of New Mexico. Studies at Hanford, Wash.,[39] demonstrated that a tracer in a glacial outwash formation could undergo lateral dispersion of 30° in only 3.5 km of travel. The extent of longitudinal dispersion was indicated by the time (approximately 140 days) required for the tracer to pass an observation well.

Libby's group has provided further examples of the use of tritium in groundwater studies.[20] It was established that the San Fernando, Calif., city well situated 2.4 km east of the San Fernando reservoir was essentially tritium free and therefore indicated no measurable recharge from the reservoir. At the city of Santa Ana, Calif., an artificial recharge operation to restore the lowered groundwater table has been in operation for some years. Tritium analyses revealed that the recharge water is moving in as fingers and streams into the groundwater rather than in a well-mixed pattern. In the Montebello Forebay near Los Angeles, a similar recharge operation has been in effect for many years. Tritium analysis revealed that 7 years after beginning operation, the recharge water had still not reached nearby wells in three major aquifers.

Tritium studies have aided in the planning of water development by the Hawaii Water Authority and have provided measurements of the average time that the rainwater, which constitutes Oahu's freshwater storage, is held up in the porous volcanic rocks.

An example of the unique value of tritium was contributed by Carlson, Thatcher, and Rhodehamel,[21] who used the gradations of tritium with depth in the Mullica River Basin of New Jersey to prove groundwater layering, a phenomenon that was required theoretically but

could not be established by conventional hydrologic techniques. Thatcher et al.[22] studied water in wadi gravels of central Arabia. Here it was determined that the water could not be more than 10 years old and therefore has no connection with any large groundwater body. Brown[23] has presented a tritium study of the Ottawa River Basin which includes an analysis of the groundwater system. Tritium has been used by several groups to establish whether thermal waters originated from precipitation or from magmatic sources. Brown and Tremblay[24] have studied flow rates in an aquifer of Quebec province where two lines of artesian wells are influenced by an escarpment. With the assumption that 15 T.U. (T.U. = tritium unit = $T/H = 10^{-18}$) was the prebomb tritium concentration, dates for down-gradient water samples on one line were calculated to be 1955, 1930, 1926, and 1916. The calculated flow velocity was 45 m/year. The wells on the second line gave a calculated down-gradient flow velocity of 110 m/year.

Brown and Meyboom[25] have studied six deep wells and selected shallow wells in Saskatchewan. Wells at 140 to 150 m depth gave water ages ranging from 26 to 48 years. A well at 9 m gave a tritium value of 754 T.U. in 1961. This concentration indicated 1958 and 1959 water on the basis of known patterns of tritium in precipitation. From this it was estimated that the infiltration rate had been 3.2 vertical m/year, which is consistent with the rate deduced from deep samples.

Accurate application of the tritium geophysics method to hydrological problems in coastal, continental, and high- and low-altitude environments of the northern and southern hemispheres is based on accurate knowledge of the tritium input function for the area of interest. Tritium in precipitation varies widely, with the concentration generally increasing with distance from the ocean and with altitude. Concentrations at inland sites in the northern hemisphere in 1963 averaged 2000 to 3000 T.U. compared with 20 to 100 T.U. for the southern hemisphere. The IAEA and World Meteorological Organization are conducting a worldwide survey of precipitation at 103 stations. Monthly samples are collected and measured for ^{3}H, ^{2}H, and ^{18}O. Tritium results are published by the IAEA.[26]

Carbon-14 has been proposed for the study of old, deep groundwaters in confined formations. The ^{14}C half-life of 5700 years extends dating possibilities back 20,000 to 30,000 years to include recharge events during recessions of recent glacial periods. Studies have been presented by Vogel[27] and by Münnich and Vogel.[28] Studies made in Arabia[22] indicated ages of about 20,000 years for water from deep wells.

Any loss of ^{14}C by exchange with other carbon isotopes leads to the assignment of an excessive age in the dating procedure. The question of exchange must therefore be resolved to place the hydrologic application of ^{14}C on a firmer base. This is the subject of an IAEA-sponsored investigation by Münnich and Roether.[29]

RESEARCH IN GROUNDWATER TRACING

Dispersion. Study of dispersion phenomena in groundwater is of interest whenever knowledge of movement of dissolved substances in the subsurface is desired. The use of artificially injected tracers to develop knowledge of permeability or groundwater velocity and direction of flow together with studies of the spread of pollution or contamination in groundwater are the major types of investigations that require an understanding of dispersion mechanics for interpretation. These studies involve introduction of a tracer and observation of the dilution that occurs during flow through the aquifer. The amount of dilution that occurs is used as an integrated index of dispersion caused by the complex physical and chemical characteristics of the system.

As early as 1887 Thiem[30] developed a method for measuring groundwater velocity by dispersing a salt tracer in an aquifer; his method was further developed by Slichter.[31] These early studies of groundwater movement, however, were often frustrated by lack of knowledge of the physical dispersive and chemical phenomena that critically affect the movement of dissolved constituents in groundwater. Concern over possible groundwater contamination from radioactive wastes has recently spurred isotope and groundwater specialists into cooperative efforts in complex research studies of the dispersion of contaminants in the ground. As a result, much of the new knowledge of these mechanisms appears in publications relating to nuclear energy.

No completely satisfactory comprehensive synthesis of groundwater dispersion theory has been developed. However, individual studies of

longitudinal and tranverse dispersion have been made by several investigators.

De Josselin de Jong,[32] Ogata and Banks,[33] Skibitzke et al.,[34] and Harleman, Melhorn, and Rumer[35] have investigated longitudinal dispersion phenomena. Ogata,[36] Simpson,[37] and Theis[38] have developed knowledge of transverse dispersion in flow through porous media. Efforts to integrate knowledge of both longitudinal and transverse dispersion in groundwater flow on a field scale have been made largely on an empirical basis by Bierschenk,[39] Newton,[40] Parsons,[41,42] Jones,[43] Inoue and Kaufman,[44] Skibitzke and Robinson,[45] and Theis.[18]

Theis has emphasized heterogeneity in geological formations as being of utmost importance in influencing the spread of contaminants in the subsurface. Without making a direct or detailed assessment of the heterogeneity of the formation through which dispersion takes place, Inoue and Kaufman[46] have developed a method for predicting the breakthrough times and concentrations of radionuclides moving in solution through the ground. However, the accuracy of the prediction varies as a function of the heterogeneity of the strata. Their prediction was closely verified in a formation having a permeability range of about 3.5 between minimum and maximum.

The Weizmann Institute is also conducting research in the dispersion of tritium during underground movement. The model of piston flow appears to be satisfactory for relatively homogeneous, confined aquifers when the length of travel exceeds the depth of the formation by a large factor. The field experience of others, however, indicates that piston flow may be the exception rather than the rule. Undoubtedly the problem requires much study, and early conclusions must be accepted with caution.

Nelson and Reisenauer[47] have developed perhaps the most advanced mathematical approach to hydrodynamic dispersion in tracer tests. They point out, however, that this approach can be used meaningfully only when applied to a system for which a detailed basic flow analysis is available. The difficulty and expense of developing such an analysis to which tracer tests may be routinely applied is very great and can seldom be justified in normal groundwater-supply investigation. In areas where industrial or radioactive wastes are discharged into aquifers, however, an elaborate analysis may be the only means of obtaining the required knowledge.

Comparative Tracer Evaluation. The literature contains many reports of tracer studies and laboratory tests of individual tracers, but few comparative evaluations of tracers under controlled conditions have been offered since the early work of Kaufman and Orlob[48] and others. The conclusions of field experiments are frequently weakened by uncertainty about the tracer performance. A critical report on the performance of nine radioisotope tracers for use in water-flooding operations in oil fields was presented by Heemstra et al.[49] They studied both breakthrough characteristics and the long-term loss of tracer under continuous recycle conditions in Roubidoux sandstone. It was established that tritiated water was the best tracer and that ^{131}I and ^{124}Sb (EDTA) were also satisfactory. Cobalt-60 (EDTA) and ^{192}Ir (EDTA) were less satisfactory but could be used if corrections for tracer delay and loss were made.

Other papers that provide useful information on tracer performance are Hours,[50] Lacey and de Lagune,[51] Halevy et al.,[52] and Knutsson et al.[53]

BOREHOLE LOGGING

Radiation logs are used in boreholes for locating and studying the characteristics of water-bearing strata. Three types of radiation logs are in common use: (1) the natural gamma log, which measures radioisotope content of rocks; (2) the neutron log, which measures hydrogen content; and (3) the gamma backscatter (gamma-gamma) log, which measures formation density.[54,55]

The contents of the naturally occurring radioisotopes of uranium, thorium, and potassium differ significantly from one rock type to another. Silts and clays generally show higher gamma activities than gravel and sand formations. Limestones and basalts, which are important aquifers in some areas, generally have very low gamma-ray activities. When electrical methods of logging are ineffective, the natural gamma log is most useful for cased holes, dry holes, or holes filled with oil-base muds and is used mainly to obtain correlation from one borehole to another.

The neutron log uses a radioisotope source of fast neutrons and measures the effect of the strata in slowing down or moderating the neutrons. Hydrogen is the most effective element for neutron moderation, and hence the neutron

log gives a signal that decreases with increasing hydrogen content. Thus, if consecutive natural gamma and neutron logs are run in a borehole, both the natural gamma signal and the neutron signal will be low where penetration of water-bearing limestone or sandstone occurs. Clays that have a high bound-water content can be identified by the high signal they give.[56]

Formation density obtained by gamma-gamma logs can be used to evaluate porosity when combined with information on fluid content and fluid density.[55]

New developments in radioisotope logging include the use of borehole neutron generators and neutron-activation logs, neutron gamma-ray spectral logs, and natural gamma-ray spectral logs.[57]

Radioactivity logging techniques as an adjunct to groundwater investigations can be particularly valuable when combined with (1) other geophysical logging techniques, including resistivity, spontaneous potential, temperature, sonic, and caliper logging, and (2) examination of drill cuttings, water-quality difference, and other available hydrogeological, geophysical, and geochemical knowledge.

Application of Radioisotopes to Surface-Water Resources

Efficient use of surface water for domestic, agricultural, industrial, and hydroelectric purposes requires accurate measurement of the amount of available water in rivers and as precipitation and soil moisture.

GAGING THE FLOW OF RIVERS

There are four principal methods for measuring the flow of water in streams, canals, and rivers. One involves the establishment of a stage discharge relationship at a gaging site, which enables the flow to be estimated from the given stage height of the river. Flow rates are also measured from the discharge-head relationship established for hydraulic structures, for example, critical-throated flumes. In another frequently used method, known as the velocity-area method, the velocity at a number of points across the cross-section is measured with a current meter, and the flow is calculated from the velocity and the area of the cross-section. Difficulty in determining the area and its possible change with time detract from this method. With large rivers the method is quite time consuming and will lead to errors if there is a change in flow rate during the measurement of the velocity. In the fourth technique, the salt-dilution method, a known amount of salt is introduced into the stream, and, at a point sufficiently far downstream to permit complete transverse mixing, the salt concentration is determined. In some countries this method is routinely used for flow rates up to 50 m^3/sec. The method is not easily applicable to muddy water, and for higher flow rates this method becomes more difficult and less economical because of the larger quantities of tracer required.

The use of radioactive material was suggested for flow gaging as early as 1922 by Joly.[58] However, it was not until artificial radioisotopes were more widely available that serious attention was given to the possibility that the higher sensitivity of detection of radioisotopes might lead to more accurate measurements. Since radioisotopes are simply another form of tracer, they have been used with the salt-dilution technique. Their use in the velocity-area method has been restricted mostly to flow in pipes because of the difficulty in measuring the cross-sectional area of open streams. The following discussion is therefore restricted to the different modifications of the salt-dilution method.

The tracer may be injected instantaneously into the stream as in the pulse or integration method. If A is the quantity of tracer injected, then the mass flow equation is

$$A = Q \int c\, dt$$

$$Q = A / \int c\, dt$$

where c = concentration of the tracer in the stream
t = time
Q = flow rate of the stream

This method consists in evaluating $\int c\, dt$ at a measuring point downstream where transverse mixing is complete.

In the dilution method the tracer having a known concentration, C_1, may be continuously injected into the stream at a known rate, q. Sufficiently far downstream to ensure complete transverse mixing, the concentration of the tracer, C_2, is measured. If Q is the flow

rate of the stream, the following mass balance is obtained:

$$qC_1 = (Q + q)C_2$$

Thus

$$Q = q[(C_1 - C_2)/C_2]$$

Hull[59] first reported the use of ^{198}Au in the pulse method. A Geiger counter was submerged in the stream to record the total observed count from the radioisotope as it passed. The counter had previously been calibrated in a vessel containing a solution of ^{198}Au of known concentration. Moser et al.[60,61] have also employed this technique using ^{82}Br, ^{131}I, and ^{24}Na and a scintillation detector. They pumped water from the stream through a counting container of known geometry and thus avoided the difficulty of placing the detector in an "infinite volume" in the stream. Clayton and Smith[62] have also investigated this method by pumping water from the stream through counting containers placed in the ground to shield them from the tracer passing in the river. This work was part of a comparison of several isotope methods. In the continuous sample method, periodic samples were taken during the passage of the tracer pulse. The integral was computed from the concentration of the samples.

Guizerix et al.[63] have a different approach to the determination of the integral. They remove water from the stream by means of a small battery-driven pump into an 18-liter container fitted with a constant-head device. The diameter of the input pipe to the container from the constant-head device may be adjusted so that the time for filling of the container embraces the time of passage of the tracer pulse. A scintillation counter is immersed in the 18-liter container to measure the mean concentration of the total sample. The same container is used for counting an aliquot of the injected solution. Both this technique and that of taking individual samples have a number of advantages over the immersion of a detector in the stream. The difficulty of placing a detector in an infinite volume is avoided, which is particularly important in shallow rivers.

The dilution method requires the constant injection of a solution of radioisotope of known concentration for a time sufficiently long to ensure unvarying concentration at the sampling point. This concentration may be determined by placing the counter directly in the stream, by placing it in a container through which water from the stream is pumped, or by taking samples.

The prime requisite for all tracer methods is complete mixing at the sampling point. A rough indication of this location may be obtained visually in small streams by injecting a dye. Guizerix employs this method to check mixing by pumping samples from points across a river.

A number of attempts have been made to estimate the mixing distance or length. Hull[64] has proposed that the mixing length, L_{mix}, is given by

$$L_{mix} = aQ^{1/3}$$

where a is 50 for center-point injection and 200 for injection at the side of the stream and Q is in cubic feet per second.

A number of subsequent experiments by other workers have shown that this formula underestimates mixing length.

Rimmar[65] assumed that vertical mixing was much more rapid than transverse mixing and proposed the formula

$$L_{mix} = 0.13C(B^2/H)[(0.7C + 6)/g]$$

where C = Chezy coefficient
B = breadth in meters
H = depth in meters
g = gravitational constant

This formula seems to give more realistic estimates of the mixing length. However, a more systematic study is required of the determination of this factor and also that for the longitudinal dispersion.

The longitudinal dispersion determines the duration of sampling and also the quantity of radioactive tracer to be used. Smith,[66] using the hydraulic radius of the stream and estimating the friction factor of the bed, reports a fairly close agreement with the theoretical dispersion calculated from Taylor's[67] formula for dispersion in turbulent flow through a pipe. Present experience shows that a few tens of millicuries of ^{82}Br or ^{131}I per cubic meter are necessary for the pulse-injection method of measuring flows of about 10 m^3/sec.

The choice of the radioisotope for flow measurements depends on half-life, type and energy of emitted radiation, maximum available specific activity, and the maximum permissible level in drinking water. Careful consideration must be given to the possible loss of tracer by adsorption, which would invalidate the method. Gold-198 should be viewed with suspicion, and losses of ^{131}I may occur. Care must also be taken with iodine to assure that it remains in the anionic form.

Each method has its advantages and disadvantages. With the instantaneous-pulse method it is difficult to measure the long tail of the pulse. With the continuous-dilution method, when the period of injection is too limited and large longitudinal dispersion occurs, it can be difficult to establish the equilibrium concentration at the sampling site; thus the pulse-injection method would be preferred. Moreover, for larger flow rates the quantity of tracer required for the continuous-dilution method would become uneconomical. Gaging of large flows would be of widespread applicability if the accuracy of 1 or 2% obtainable at present with flows up to 100 m^3/sec could be retained. The quantity of radioisotope necessary for large flow rates may be uneconomical and may also present difficulties in the handling of heavily shielded isotope containers. One answer may be an extension of the chemical concentration procedure that has been examined by Moser. He has precipitated silver iodide from the water samples and counted the precipitate. Guizerix has shown that a factor of 10 increase in sensitivity can be obtained by concentrating the radioisotope from the solution onto an ion-exchange resin. The ultimate answer to the measurement of flows greater than 1000 m^3/sec may lie in the use of tritium, which does not present the problems of shielding and is relatively cheap. Consideration should also be given to injecting inactive tracer and measuring its concentration by radioactivation or by the radiorelease method. In the latter a suitable inactive tracer releases radioactive ions from a radioactive metal.

MEASUREMENT OF SNOW DEPOSITION AND SOIL MOISTURE

The mass of snow deposited in a catchment area is estimated by choosing snow courses about 200 m long, which are found by experience to be representative of much larger areas, and by weighing samples taken every 20 m along the course. The number of these snow courses need not be large to cover a wide area. For example, most of the spring runoff from the Andes into Chile and Argentina is predicted from only five snow courses, and it is possible to predict the runoff with an accuracy better than 20% in 99% of the measurements.

For a prolonged series of measurements, one representative point on a snow course can be chosen. Measurements at such points are extremely difficult and hazardous even when helicopters are used; hence a self-contained instrument that continuously measures and either stores the information for periodic collection or transmits it to a central accessible station is highly desirable.

Gamma-transmission gages have been developed for this purpose. In one design a 40-mc ^{60}Co source is mounted at the lower end of a heavy lead tube inserted in the ground with the Geiger counter detector mounted 5 m above the ground.[68] The accuracy of this instrument is better than 5% up to the water equivalent of 110 cm.

Fundamental research on the hydrological characteristics and thermal qualities of mountain snowpacks is being conducted with gamma-backscatter density gages.[69] It is hoped to correlate density variations and the presence of ice lenses with the rate of melting to predict runoff more accurately and also to assess the effect of vegetation in delaying melting and thus preventing floods.

The amount of water in soil and rock strata can be measured with neutron-moderation moisture gages.[70] These instruments have a radioisotope source of fast neutrons, such as radium-beryllium, a slow-neutron detector, and a means of indicating the neutron counting rate. A BF_3-filled proportional counter is used as a detector, and the counting rate is measured with a portable battery-operated scaler or rate meter. Fast neutrons from the source are slowed down in the soil mainly by collisions with hydrogen atoms; hence the number of slow neutrons produced is a function of hydrogen or water content. Measurements are made both on the surface and at depth.

With a source activity of about 10 mc, moisture content in the range 1 to 50 vol.% can be measured with an accuracy of about 1% in

counting times of about 1 min. The instrument determines the water content in a cylindrical configuration that is about 6 in. in diameter and has a length equal to the distance between source and detector.

If the water content is required in weight percent, an independent measurement of the soil density is required. This is accomplished with a gamma-backscatter density gage that gives an accuracy of 1% in a measuring time of about 1 min.

MOVEMENT OF SAND AND SEDIMENT IN RIVERS AND ESTUARIES

Sediment transported by rivers and subsequently deposited at points where turbulence and velocity are insufficient to transport it further is a major problem because the silt gradually accumulates in reservoirs, navigable rivers, and estuaries. Costly dredging schemes are often necessary to keep rivers and estuaries navigable, and sediment deposited in reservoirs limits the life of dams in some areas by reducing the storage volume. In some instances the storage volume of reservoirs has been reduced by a factor of 2 in 10 years. Sediment content of rivers also provides an integrated measure of the effectiveness of land conservation methods. Thus an accurate assessment of the rate of transport of sediment is required.

Sediment is transported both in suspension and along the bed of the river. The mass in suspension is smaller than the moving bed load; however, because of the higher velocity of the former, the proportion of sediment transported in suspension is often greater than 75%. The maximum concentrations reach as high as 200 g/liter, but the concentration range of most interest is from 1 to 50 g/liter.

A portable instrument[71] for analyses in this range has been constructed; it uses the preferential absorption of 22-kev gamma rays from ^{109}Cd. With a 2-mc source, an 8-cm separation between the source and scintillation detector, and a 15-min counting time, this instrument has an accuracy of 20% at a concentration of 1 g/liter; better accuracies are obtained at higher concentrations.

Transport of silt, sand, and pebbles as bed load in rivers, irrigation channels, and estuaries is determined with artificial silt labeled with a gamma-emitting radioisotope.[72] The movement of this silt along the bed of a river or estuary is followed with a scintillation or Geiger counter detector towed along the bottom on a sledge. The counting rate is indicated on a rate meter in the towing boat.

The silt is labeled with either reactor-irradiated glass particles containing a specific material for activation or a radioisotope adsorbed onto the surface of the grains. Radioisotopes used include ^{182}Ta, ^{192}Ir, ^{51}Cr, ^{46}Sc, ^{65}Zn, and ^{60}Co. The labeled sand is matched in particle size distribution and specific gravity with the sand to be followed. The activities range from a few tens of millicuries to 20 curies. Movement of the tracer has been followed up to several miles.

All studies to date have shown the direction and rate of movement of the labeled particles. Attempts are being made to obtain a quantitative estimate of the mass rate of flow of bed load using methods similar to those outlined above for stream gaging.[73] A good review of this subject has been made by Griesseier.[72]

Pollution Studies

As countries become more industrialized and centers of dense population become more numerous, pollution and quality of water supplies assume an even greater importance.

WATER QUALITY

The impact of nuclear techniques on pollution studies has been relatively less than in other areas of hydrology because the existing methods of water analysis are generally satisfactory. Sensitive conventional techniques, e.g., emission spectrography and polarography, are available for trace-element analysis. Nevertheless, as industrial requirements become more demanding, as for example in the manufacture of transistors, and the physiological role of trace elements in water becomes better understood, the application of sensitive nuclear techniques is likely to increase.

A system for complete chemical analysis of water by neutron-activation analysis has been developed by Blanchard et al.[74] They combined gamma-ray spectrometry with simple elemental-group chemical separations based on the traditional qualitative analysis scheme to determine both the major constituents and

the following trace elements: As, Br, F, Cu, Mn, Rb, Sr, and Zn. Accuracy for trace elements was superior to the spectrograph. This analytical scheme could be made economically competitive with a sufficient volume of work. Lyon and Reynolds[75] have detected 20 elements in tap water by neutron activation.

The problem of detergents, fertilizer residues, and pesticides with their decomposition products is becoming acute in groundwaters and surface waters of certain areas. Detergents in groundwaters can be traced with a detergent molecule labeled with a radioactive atom. Some pesticides may be toxic at extremely low concentrations; unfortunately methods of accurate analysis at these concentrations are still in the research stage. A promising approach is the use of sensitive isotope techniques based on the dilution of labeled DDT, dieldrin, aldrin, etc., by the inactive pesticide in the water.

EFFLUENT DISPOSAL

Several tests for effluent disposal with pipelines, treatment tanks, and hydraulic models have been described. Radioactive materials have been used in evaluating the quantities of radioactive waste that may be discharged safely into a river. However, there is an increased interest in the use of radioisotopes for tracing sewage effluent. Studies have been made of the flow characteristics of an anaerobic sewage-treatment plant[76] and of the tracing of sewage discharged into the sea.[77] Putman et al.[78] compared the use of radioisotopes and an easily recognizable bacteria for the labeling of sewage. Labeled-sewage techniques are now helping to establish the proper design of effluent pipelines to guard against beach pollution. Earlier work gave qualitative information on the flow patterns, but Berg and Somer[79] have extended the method and are able to predict the concentrations of sewage water to be expected from long-term buildup at different points. They make a point injection of ^{82}Br as ammonium bromide and assume that a continuous injection constitutes an infinite number of identical short term injections. A chart prepared by integrating the activity concentration with time shows the distribution of sewage concentration to be expected from a sewage outlet with a given flow.

A Danish group has further developed the method,[80] particularly in presenting the data in a form that demonstrates the most economical disposal in terms of effluent-pipe length and sewage-purification factor. The two extremes are a long pipe discharging unpurified effluent far from shore and a short pipe discharging highly purified effluent. In practice, injections of ^{82}Br are made at three different points corresponding to varying effluent-pipe lengths. A diagram is then prepared showing the total cost for the most economic disposal method. Plans are being made for equipping a boat specifically for this type of investigation, and it is hoped to record all the data on punched tape, which can be fed into a computer when the boat returns to harbor.

Conclusion

Isotope techniques are already being used in many branches of hydrology, and they should be considered available tools capable of providing complementary and in some cases unique information to the hydrologist. In the solution of certain hydrological problems, isotope techniques can be more advantageously employed than other methods, and they provide shortcuts to the desired goals.[81] The potentials of isotope techniques should be given consideration in the planning of investigations leading to the rational development of water sources.

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Some Applications of Radioactive Tracers in Civil Engineering

By M. del Val Cob, A. Plata Bedmar, and J. M. Serratosa Marquez*

Paper 497 (extracted)

Three road-surfacing problems were solved with radiotracers: determining the inclination of strain gages in concrete slabs, locating metal parts used to measure the settling of a road surface, and estimating uniformity in ligand spraying.

For strain measurements gages are inserted in the concrete slabs used for road surfacing in a plane normal to the road axis. Although the location and inclination of these devices are originally known, they change while the concrete is being spread to form a slab. The inclination with respect to the main tension axis (road axis) must be measured with an accuracy of at least ±5% to make appropriate corrections in experimentally obtained strain values. This correction was made by fixing two small $Na^{131}I$ sources to the strain gage and then measuring the radioactivity from the surface with an appropriately collimated scintillation counter (Fig. 497-1).

In a series of experiments, strain gage chambers were buried more deeply than in field conditions, and inclinations of 0, 3, 4, 10, 15, 20, 25, 30, and 35° were simulated.

When the chamber is vertical, the two sources are colinear with respect to the vertical axis, and counting rate is maximum. As the inclination angle changes, two counting-rate peaks corresponding to the two radioactive sources result (Fig. 497-2). For each inclination step the separation between peaks is equal to the distance between the vertical planes on which the two sources are located. From this value and the value of the distance between the

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two fixed sources (7 and 11 cm were tried), the angle of inclination was determined. Accuracy was generally better than ±1%.

A complete setup was constructed for actual determinations on the road. It is now routinely used.

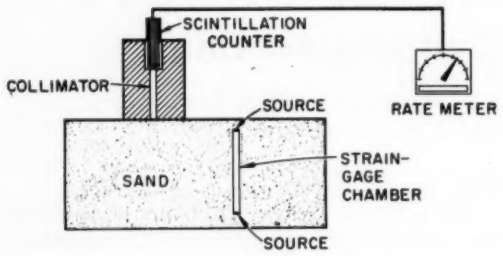

Fig. 497-1 Experimental setup to determine the inclination of a buried strain gage.

The settling of a road surface is determined with a mechanical device that indicates the difference in distance between two rectangular metal plates. One of these plates is fixed to the road bed and the other in the road surface layer. The center of the road-bed plate needs to be located after the road coatings have been applied. Locating its center by drilling through the road is time consuming and difficult. Hence about 100 to 200 μc of $Na^{131}I$ covered with Araldite was applied to the center of the plate before it was fixed to the roadbed. A scintillation counter with a lead collimator located the plate. The center of the plate was found as follows: the zone was scanned, and a straight line was drawn on the road along the line of maximum activity in the direction of the collimator slit. The slit was then turned 90° in the same plane, and the road was scanned again along this new line. A second line was drawn through the area of maximum activity. The intersection of these two lines marks the center of the plate and simplifies the drilling operation.

Conventional methods of estimating the uniformity of sprayed binders, not always satisfactory, depend on sampling of paper strips spread on the road. In these experiments road conditions were simulated, and ^{82}Br-labeled olive oil was dissolved in the binder in a search for more satisfactory methods. Layers of different activity of radioactive binder and gravel were spread on top of macadam, and radioactivity was measured with a lead-collimated scintillation counter connected to a rate meter. Laboratory experiments were aimed mainly at studying the possible sources of error that might affect measurements and at finding the optimum experimental conditions for applying the method to actual road measurements. Some of the possible sources of error investigated were (1) influence of the amount of gravel on the counting rate, (2) thickness of the gravel and binder layer, (3) lack of radioactive homogeneity in the labeled asphaltic binder, and (4) contribution to the counting rate of zones outside the collimator as a function of lead thickness.

In a proposed method for road measurements based on these experiments, the following conditions were recommended: (1) the scintillation NaI(Tl) phosphor should be as large as possible; (2) the lead collimator should have a large aperture to measure large areas of the radioactive layer; and (3) the counter-to-surface distance should be as short as possible.

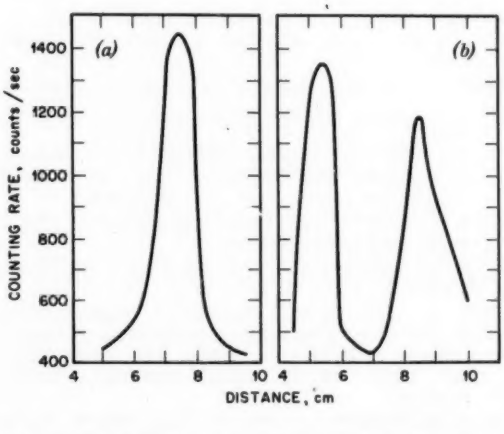

Fig. 497-2 Counting rates for actual inclination of (a) 0° and (b) 30°. Inclination determined for (b) was 28° 7′. Sources were 7 cm apart. Data have been replotted.

A lead collimator was built for measurements on the road. It has an inner conical shape and covers a measuring area of 314 cm^2. With this collimator and a 2- by 2-in. scintillation crystal, determinations of asphaltic binder were made accurately with a specific activity of <0.2 μc/kg.

Pipeline Wear in Iron-Ore Transportation

By H. L. Tavares, P. E. Aun, A. A. Maestrini, and M. T. Magalhães*

Paper 114 (abstracted)

At the request of a metallurgical and mining company, we determined the wear of a pipeline used for transporting iron-ore fines in aqueous suspension by activating a section of a pipe and measuring the wear.

Wear is roughly the sum of corrosion and abrasion. A loop for the circulation of the slurry was built and a radioactivated steel tube was inserted and its wear studied. Since wear from the tube was proportional to the total count of ^{59}Fe present in the slurry, wear was determined by adequate sampling and by use of a well type scintillation detector [NaI(Tl) crystal, 1¾ by 2 in.].

Preparing and counting a standard with a known mass of the same steel irradiated under the same conditions as the tube, we related the total count obtained to the weight of worn material. This standard was also used for correcting the decay time of ^{59}Fe.

Three steel tubes of different composition were irradiated, two of them in a TRIGA Mark I reactor at 30 kw and one of them in a swimming pool reactor at 2 Mw.

We found that wear rate decreases with transportation time; this could be due to the variation of the grain size and shape during the experiment. Further tests and photomicrographs demonstrated that the iron-ore grains were ground and rounded during transportation. Hence the pipeline life will increase in the direction of flow, or the tube will become more worn nearest the point at which the ore is introduced. Grain size and shape variation were studied in dry and wet ground iron ore. We found that wear increases with increasing particle diameter and that finer iron ore and wet ground iron ore cause less pipe wear than dry ground ore. Generally speaking, in all experiments for relatively long transportation times, the wear rate ranged from 0.001 to 0.003 mg/(cm^2)(hr). The rate of decrease in thickness for a number of transportation times varied in six experiments from 0.04 to 0.40 mm/year. From the rates of decrease in thickness found for specific types of ore with pelletizing characteristics, a preliminary design of an 18-in.-diameter pipeline was made, and ore transportation costs were determined. A study showed that this pipeline could carry ore three times faster than a railroad connecting the same points. On the basis of this study, a pilot pipeline (8 in. in diameter) is being built.

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Measurement of High-Dose Gamma Fluxes by the Conductivity of Cuprous Oxide

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Paper 815 (abstracted)

Cuprous oxide with a coarse crystalline structure was produced from copper plates (1 by 20 by 80 mm) by a special oxidation process. The specific electrical conductivity of this material is similar to that of single crystals. In fine crystalline specimens the conductivity increases proportionately with the number of crystals per unit area. This behavior is the result of a higher concentration of crystal defects on the grain surfaces. Under the influence of gamma and X-ray irradiation, the conductivity is increased; the lower the initial conductivity of the material, the greater the relative effect. The results obtained showed a linear dependence and proved coarse crystalline Cu_2O to be most suitable for high-gamma flux determinations.

One curie of ^{137}Cs and 1000 and 2000 curies of ^{60}Co were used as sources at a constant temperature (±0.01°C). On the basis of results of the measurements with the ^{137}Cs source, the relation between grain size per unit area and the relative decrease in resistance was examined. It was shown that coarse crystalline samples were considerably more sensitive to radiation than fine-ground samples. With cu-

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prous oxide samples the gamma flux of sources of different strengths was determined, and the dose rate of a TRIGA Mark II reactor was measured in places accessible only with great difficulty to other detectors. The electronic circuit for these experiments was a very simple resistor-capacitor setup connected to a recorder and a battery.

It was concluded that small cuprous oxide detectors, either as single crystals (10 mm^2 in surface area) or as polycrystalline samples (several square millimeters in surface area), can be used for measuring intense gamma radiation in places where access is difficult. From the proportionality of conductivity to dose and the measured resistance difference in cuprous oxide before and after irradiation, the dose can be determined by a single calculation.

Enrichment of Low-Level Tritium by Thermal Diffusion for Hydrological Applications

By B. Th. Verhagen and J. P. F. Sellschop*

Paper 465 (extracted)

The University of the Witwatersrand two-stage thermal-diffusion tritium separator described in this paper has the advantages of good reproducibility, short running times, and a relatively high yield. The difficulties of feeding the system and sampling and purifying the product have been overcome. The apparatus is proving to be a flexible and useful tool in a tritium laboratory.

Description of Separation Plant

The tritium separator was designed on the basis of the theory of Jones and Furry.[1] A brief description of the separator is given in Ref. 2. The system consists of two columns. The upper, or first stage, is a concentric-tube column, and the lower, or second stage, a smaller hot-wire column. Diagrammatic sections are shown in Figs. 465-1 and 465-2.

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Tube *a* in Fig. 465-1 is heated by a nichrome heating spiral, *h*, operating on 380 volts alternating current. On expanding, this tube slides through gland *s*. It is water cooled at either end at *r* to protect the neoprene seals. Tube *b* is the cold wall; cooling water circulates between it and water jacket *c*. Feed gas is introduced through tube *i* which enters the column at its midpoint, and gas can leave the column at *o*. The connections to the second stage are at points *g*.

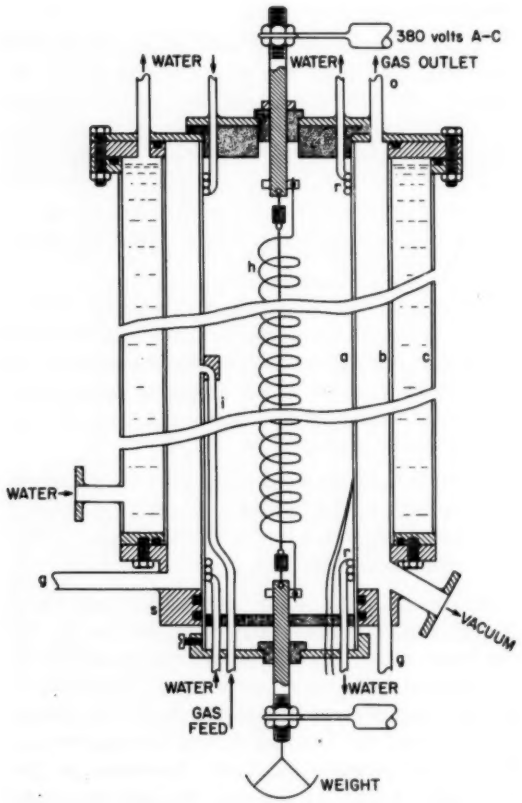

Fig. 465-1 Section of first stage concentric tube column. (See text for explanation of letters.)

The hot wall *a* in Fig. 465-2 is a d-c heated tungsten wire attached to a sliding-seal electrode, *f*; *b* is the cold wall; and *c* is the water jacket. Connections to the first stage at the top and to a purification/extraction system at the bottom are at *g*.

Table 465-1 gives the principal dimensions and operating conditions of the two columns.

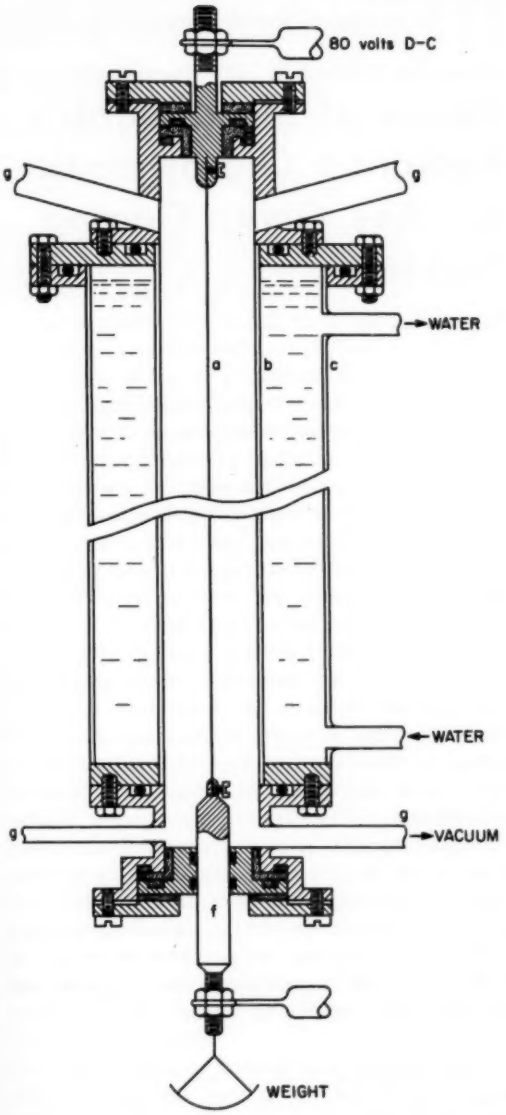

Fig. 465-2 Section of second stage hot-wire column. (See text for explanation of letters.)

Some Results from Calibration Experiments

Figure 465-3 shows a typical curve from an enrichment run in which the column was fed with hydrogen spiked with tritium to 4.8×10^5 T.U. (T.U. = tritium unit = $T/H = 10^{-18}$). The resulting enrichment was observed with the flow ionization chamber mounted in the purification line. The initial curved part of the graph represents the period in which the first-stage column separates material faster than it is withdrawn by the second stage. Eventually steady-state conditions are reached, and the enrichment becomes linear with time. The curve shown in Fig. 465-3 can be used as a calibration curve to find the enrichment after a certain running time because this curve is reproduced closely in independent runs.

Table 465-1 PRINCIPAL DIMENSIONS AND OPERATING CONDITIONS OF THE TWO THERMAL-DIFFUSION COLUMNS

	Concentric tube column	Hot-wire column
Length, m	7.20	2.75
Radius of hot wall, cm	6.30	0.02
Radius of cold wall, cm	8.76	1.50
Volume (cold), liters	80	2
Temp. of hot wall, °K	600	1200
Temp. of cold wall, °K	300	300
Power consumption, kw	18.5	1.66
Equilibrium separation factor (theoretical)	23.2	432
Operating pressure, atm	1	1

Although the activity of 4.8×10^5 T.U. still strictly conforms to the assumption in the design formulas[3] of the system, i.e., the concentration of the desired isotope shall be $\ll 1$, a few short runs were made with lower activities to check the validity of the calibrations to values of interest (1 to 1000 T.U.). The results showed that there is no significant dependence of enrichment on concentration and therefore that the calibration curve is valid for all concentrations of interest.

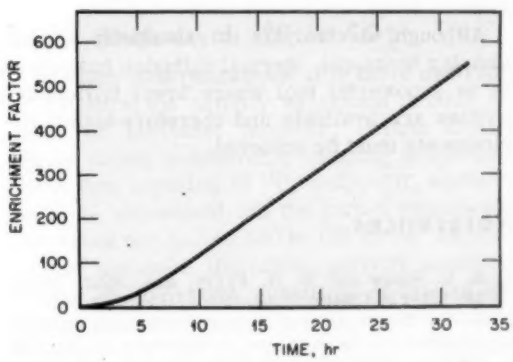

Fig. 465-3 Typical enrichment curve for whole plant.

Detection

The low-level counting system in this laboratory consists of a 500-ml proportional counter with an anticoincidence ring and 4 in. of 60-year-old lead shielding. The full spectrum count with the anticoincidence system is 12 counts/min. With a window set at the maximum energy of the beta particle, E_{max}, for tritium, the count is reduced to 6 counts/min.

The hydrogen obtained from the column is reacted with acetylene to produce ethane.[4] Thus two molecules of hydrogen are incorporated in each molecule of counting gas, and a 1-liter yield from the separator can be held in the counter.

At 50 times enrichment the minimum detectable activity of the system lies at about 5 T.U. This can be extended down to 1 to 2 T.U. by longer enrichment runs. A commercial screen-wall counter would reduce the minimum detectable activity to 0.5 T.U.

Discussion

The separation system described above shows good reproducibility and can give a range of enrichments to suit the activity of the feed material. Withdrawal experiments correspond well with theory and provide a convenient means of collecting enriched material in any required amount.

The production of enriched material at higher enrichments (>100) becomes prohibitively small. A redesigned system in which the values of H (transport coefficient of thermal diffusion in grams per second) for both stages would be larger and the lengths smaller would produce material at enrichments of about 100 at much greater rates and in less time.

Although electrolysis is essentially a much simpler technique, thermal diffusion has proved to be a powerful tool where lower tritium activities are available and therefore higher enrichments must be achieved.

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Choice of Most Suitable Radiation Characteristics for Any Given Gaging Problem

By P. Platzek and A. C. Meyer*

Paper 828 (abstracted)

Most gaging is directed toward detection or measurement of mass differences rather than the determination of an average mass. In this respect level indication may be considered a gaging problem as well as thickness or density (difference) measurements. When secondary effects, such as heterogeneity of radiation, scattering, source dimensions, and detector response, are neglected, beta as well as gamma radiation can, to a first approximation, be expected to obey the same exponential absorption law. Simple theoretical considerations then show that the average object mass alone determines the choice of the optimum radioisotope (mass absorption coefficient of the principal radiation). Consequently the required source strength depends on the minimum mass difference to be detected (Δx), minimum measuring time (t), geometry and detector efficiency, and accuracy wanted ($\Delta N/\sqrt{N_x}$), where N is total count, i.e., counting rate times time; N_x is total count with object measured in place; and ΔN is count variation caused by $\Delta x = N_x - N_{x+\Delta x} = N_{x-\Delta x} - N_x$.

When signals are defined as total count, formulas can be derived to show the intercorrelation of all important variables and parameters from which the optimum radiation can be chosen. A nomogram was constructed to represent optimum as well as nonoptimum conditions. Experimental data show to what extent the simplified assumptions are valid. They justify the proposal to consider all mass-difference measurements by means of high-energy radiation as a general problem that can, in spite of the large variation in objective, always be approached in essentially the same way.

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Gas-Phase Methods Operating at Atmospheric Pressure for Analysis of Soft-Beta Emitters

By S. Mlinko* and T. Szarvast

Paper 855 (extracted)

Gas-counting and liquid-scintillation methods are almost exclusively employed to measure the radioactivity of organic compounds labeled with soft-beta-emitting isotopes, e.g., ^{3}H, ^{14}C, and ^{35}S. The most exact analysis can be made in the proportional region by an internal gas counter, which has been reported by Bernstein and Ballentine.[1] However, filling such counters requires cumbersome vacuum systems. A further complication may also arise if the analysis of carbon content of compounds is necessary because such determinations are generally made with manometric methods in a carefully calibrated volume of the vacuum system.

In our laboratory an effort has been made to eliminate the difficulties accompanying the traditional analysis of radionuclides of labeled organic compounds by developing simple new methods operating exclusively at atmospheric pressure instead of using inconvenient vacuum systems. Atmospheric operation can be achieved by adapting suitably constructed counter systems to the classical combustion methods of organic microchemical analysis. If highly precise activity results are to be obtained, the method of choice must meet the following conditions:

1. Radionuclides to be analyzed must be transformed into simple inactive gaseous compounds possessing favorable properties for proportional counting.
2. Any errors in activity from incomplete transformations, isotope effects, isotopic exchange, or cross contamination between samples must be avoided.
3. Activity results must be relative to weighed quantities of original compounds rather than to the volume of gas being measured.

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On the basis of these general principles, simple new methods have been developed for the quantitative determination of ^{14}C in methane (Ref. 2), ^{14}C in carbon dioxide (Ref. 3), ^{3}H in ethane (Ref. 4), and ^{35}S in hydrogen sulfide (Ref. 5).

Analysis of ^{14}C in CH_4

The active sample being investigated is weighed into a platinum boat, ignited in a combustion tube over cobalt oxide catalysts with air, and flushed with nitrogen for 15 min. The carbon dioxide formed is frozen out in a liquid-air trap and then converted into methane over nickel catalyst with hydrogen. The methane formed is frozen out in the trap of the counter at −215°C with liquid air under reduced pressure. The losses of methane caused by the partial pressure at −215°C are equivalent to less than 0.3%.

The methane is expanded into the counter, which is brought to atmospheric pressure with inactive methane. With a two-way trap, combustion and hydrogenation can be performed alternately; thus a determination can be completed in 35 to 40 min. Decontamination of the counter in a gas stream takes 5 min.

For wet combustion Van Slyke's reagent is used, and the apparatus is connected to the combustion tube.

The effective counter volume and the geometry are determined with standard preparations of known activity. For background determinations the counters are filled with inactive methane. Relative errors in activity determinations by both dry and wet combustion are less than ± 0.3%.

Rapid Analysis of ^{14}C in CO_2

In many determinations it is more desirable to measure the activity as carbon dioxide instead of methane; however, carbon dioxide shows rather unfavorable counting properties. Therefore counting of ^{14}C in CO_2-CH_4 mixtures is more convenient, but the partial pressure of CO_2 must not exceed 100 to 160 torrs. Because of the pressure limitation, activity measurements of equivalent quantities of labeled organic compounds as CO_2 will require counter volumes 5 to 10 times larger than measurements as CH_4. In spite of these disadvantages, carbon dioxide is frequently employed because

it can be transferred directly into the counters after combustion.

In contrast to other methods, in our experiments the carbon dioxide is frozen out from the flushing nitrogen gas under atmospheric pressure. Catalysts of high oxidative activity ensure the combustion of organic compounds in air.[6]

PRINCIPLE OF DETERMINATION

The sample is ignited in air over Co(II-III) oxide catalyst. The excess of oxygen and oxides of nitrogen are eliminated by copper. Water formed during combustion is quantitatively absorbed by an Anhydrone tube, and the carbon dioxide is frozen out in the trap of the counter. The carbon dioxide is expanded and flushed quantitatively with methane into the sensitive volume of the counter, and the activity is measured in the proportional region. For wet combustion Van Slyke's reagent is used, and the apparatus is connected to the combustion tube. The relative error of the determinations is less than ±0.3%. The counting efficiency is about 96%. The plateau of the counter extends over 350 volts with a slope of less than 0.5%/100 volts. Counting is done at 3000 volts.

Determination of ^3H in C_2H_4

Tritium incorporated into hydrocarbons has the advantages of gas counting and is less subject to isotopic exchange. However, hydrocarbons containing tritium from the decomposition of water do not give quantitative yields, and incomplete reactions may cause isotope fractionations. In our experiments both of the hydrogen atoms of a water molecule were transferred quantitatively into the hydrocarbon.

PRINCIPLE OF THE METHOD

The labeled compound is ignited in a stream of oxygen over platinum catalyst. The resultant water is frozen out and then reduced over zinc metal to hydrogen. The hydrogen is mixed with ethylene and catalytically converted to ethane over a platinum catalyst.

The ethylene-ethane mixture is frozen out in a trap permanently connected to the counter, and the activity of the gases is measured in the proportional region. Memory effects have been reduced by using a combustion tube, reducer, and catalyst tube made entirely of quartz.

Activity values are related to sample weights, and results are corrected for the memory effect of inactive combustions. A reproducibility of ±0.3%, relative, can be achieved.

The ethylene gas used for hydrogenation must be completely free from oxygen; otherwise water may form on the surface of the platinum catalyst.

The ethane-ethylene mixture gives satisfactory counting characteristics. The plateau is about 850 volts long with a slope of less than 0.3%/100 volts.

By the methods discussed, separate activity measurements of tritium and radiocarbon from a doubly labeled organic compound can also be achieved. Tritiated water and labeled carbon dioxide formed during combustion are separately frozen out in two successive traps cooled with dry ice and liquid nitrogen, respectively. Their further transformation for measurement has been described above.

Analysis of ^{35}S in H_2S

For the analysis of radiosulfur, H_2S is counted at atmospheric pressure in the proportional region. Sulfur in organic compounds can be transformed quantitatively into H_2S by reaction with hydrogen over a platinum catalyst at 900°C. Although hydrogen sulfide shows somewhat unfavorable counting characteristics, as does carbon dioxide, about 12 vol.% may be present in the counters as hydrogen sulfide without any loss in counting efficiency. The counters operate at about 3000 volts, and the plateau extends over 400 volts with a slope of less than 0.5%/100 volts.

Since hydrogen sulfide can be transferred quantitatively into the sensitive volume of the counter, a counting efficiency as high as 96% can be obtained.

PRINCIPLE OF DETERMINATION

The organic sulfur compound being analyzed is pyrolized in a flowing hydrogen atmosphere with a platinum catalyst, and the hydrogen sulfide formed is frozen out by liquid nitrogen in the trap of the counter. Halogens can be removed in an absorber containing calcium hydrosulfide and calcium chloride. The hydrogen sulfide is expanded and flushed quantitatively with methane into the sensitive volume of the counter. The relative error of determinations is less than ±0.3%.

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Section

IV

Isotopes and Radiation Technology

Process Radiation Development

Americans at Geneva reviewed developments of the past 6 to 8 years and described U. S. work on food irradiation and chemonuclear methods and the existing and potential uses of radioisotopes in process irradiation. Scientists from other countries, however, either reported data on basic research or discussed the techniques and economics of radiation processing of chemicals. The nuclear reactor and chemical processing were emphasized by both groups. One worker in this field observed that reactions having G values between 100 and 10^6 are suited to his reactor system and those above 10^6 are suited to ^{60}Co.

In this section a paper by Machurek, Dietz, and Stein, the only paper that describes the use of radioisotopes in radiation processing, is reviewed in detail. Also included is an abstract of a report on chemonuclear processing which shows the progress of recent U. S. work in this related field. This section also includes a condensed version of a German proposal for using power reactors for chemical production.

From the economic point of view, some reactions are more outstanding candidates than others for radiation processing. Previous issues of this Review have reported some of the Division of Isotopes Development work in this area,* and the Geneva paper of Machurek et al. describes a number of others. Cracking of crude oils is one of these promising reactions. A Russian paper on radiation-thermal cracking in fast-electron and in mixed gamma and neutron fluxes is summarized, and, because basic data are essential for the expansion of this field, abstracts of an Italian study of radiation effects on crude oils and a method for tracing chemical changes of fluids under gamma irradiation are given.

Isotopes Radiation Technol., 1(1): 1-26, 78-83 (Fall 1963); 1(2): 189-95 (Winter 1963-1964); 2(2): 149-57 (Winter 1964-1965).

Current Status and Future Prospects for Commercial Radiation Processing

By Joseph E. Machurek, George R. Dietz, and Martin H. Stein†

Paper 198

In the past 8 years, we have witnessed the development of a growing number of applications in commercial radiation. To regard radiation processing as an industry is premature; however, the clear beginnings of industrial commercialization of the use of radiation are at hand.

Radiation Sources

Ionizing radiation for process application is obtained from either isotopes or machines. Cobalt-60 and ^{137}Cs are leading contenders in the former category, and low- to intermediate-range (0.3 to 3.0 Mev) electron accelerators now constitute the mainstay of the machine industry. The capabilities and characteristics of some radiation sources are evaluated in the following paragraphs.

COBALT-60

A chief advantage of this reactor-produced isotope lies in its inherent ease of fabrication, containment, and handling. Its relatively high gamma energy (1.33 and 1.17 Mev) and pene-

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trability make it ideal for bulk processing. These factors, coupled with its 5.3-year half-life and its availability in large quantities at a reasonable cost, make ^{60}Co the leading choice of isotope radiation sources for the next few years.

CESIUM-137

Also a gamma emitter (0.67 Mev), ^{137}Cs is available from the separation of gross reactor-produced fission-product waste. The advantage of its 30-odd-year half-life (about six times that of ^{60}Co) is somewhat offset by its lower gamma yield and penetrability, which necessitates a curie requirement of four to five times that of ^{60}Co. Wide-scale use of cesium as a processing tool depends on achieving lower production and encapsulation costs than those presently possible. These costs are within the capability of proposed separation facilities.

A recent study conducted by the General Electric Company under contract to the AEC indicates that purified encapsulated fission products such as ^{137}Cs could be produced at about 10 cents a curie with a production throughput of 10 Mc per year.[1]

STRONTIUM-90

Like cesium, 28-year ^{90}Sr is obtained from the chemical separation of fission products. Strontium, a beta emitter, is limited in industrial processing because it has low penetration and must be encapsulated in thin-walled materials. Consequently it is more suited to liquid or gas applications, such as in high-pressure vessels, where most of the energy can be absorbed more conveniently and efficiently, with the result that pressure vessels may be smaller and less expensive. Strontium-90 is not well suited for the irradiation of bulk solids, nor can it now compete economically with machine radiation for the processing of thin films and fabrics.

SODIUM-24

This source is available only in conjunction with a liquid-sodium-cooled reactor. The irradiation technique with ^{24}Na differs from that with other isotope sources in that the sodium must be continuously "recharged" by the $^{23}Na(n,\gamma)^{24}Na$ reaction as a result of its reactor cycling. Its 15-hr half-life does not present a problem, but a low specific activity and low dose rate result in an increase in dwell time and in source inventory. Its highest energy gamma of 2.75 Mev makes it an excellent source for the processing of thick, bulky items. Transportation of the product to and from the reactor site may present economic difficulties.

An advantage not to be overlooked is that the combination of existing power reactor and isotope source could provide radiation energy at minimum cost. A case in point involves the sodium-cooled Nuclear Power Facility at Hallam, Nebr. In January 1964 the Governor of Nebraska announced a resolution of the Board of Directors of Consumers Public Power District, Hallam, Nebr., that the gamma "energy source for industrial utilization would be made available at the lowest possible cost."[2] The radiation charge would not include amortization of the power plant but would include the exceptionally low costs of incremental heat losses.

SPENT FUEL ELEMENTS

Although spent fuel elements were once considered a logical choice as a process radiation source, their use has largely been discounted. Control of the radiation field is difficult because of the variance in size, shape, specific activity, useful life, and mixed spectrum. The use of spent fuel elements as a food-irradiation source has not been authorized because of small but measurable activity induced in the food. Private commercial use is therefore unlikely at this time.

ACCELERATORS

Very intense beams of high-energy electrons can be produced by particle accelerators that convert conventional line power to electron power with good efficiency. The greatest advantage of the electron beam is the very high-intensity radiation that can be produced. The radiation intensity and output available from some of these accelerators exceeds by orders of magnitude that from any other sources of radiation that might otherwise be considered for process radiation. An impressive number of accelerators specifically developed as electron-beam sources are now in use in this country and abroad.[3] Most of the electron-beam accelerators used today (0.3 to 3.0 Mev range) are limited by their relative lack of penetration. Higher energy machines (linear accelerators) provide an electron beam of higher penetration but are significantly higher in cost and lower in reliability. Electron beams with energies up

to 10 Mev (approximately 5 cm penetration) do not produce any practical difficulties with regard to induced activity.[3]

X RAYS

Electron-beam conversion to X rays is an alternative solution to the penetration problem and offers advantages of high dose rates and partially directional beaming. Although adequate in laboratory use, X-ray units for practical commercial processing have not yet been demonstrated.

CHOICE OF SOURCE

Comparisons between isotopes and accelerators are largely invalid because of oversimplification. For example, a comparison of the superiority of machines over isotopes based on kilowatt output is misleading. The obvious advantages of isotopes are exceptional reliability, low maintenance, and simplicity of operation; those of accelerators are ultrahigh dose rates, flexibility of operation, and directional output. There are several situations where only one source of radiation can logically be used, such as the use of isotopes in processes requiring high-pressure reaction vessels or the use of electron accelerators for the production of thin films or fabrics.

For the most part the choice of a source for processing is dependent on the more subtle characteristics of both the application and the source. For instance, the fact that an isotope cannot be "turned off" is often cited as a disadvantage. However, this characteristic becomes a distinct advantage for chemical production processes that must reliably operate on a continuous 24-hr basis. The choice of the source can be made only with regard to the demands of a specific process.

Existing Commercial Applications

The total annual sales volume of radiation-processed products in the United States is conservatively estimated at $70 million and is growing rapidly. Several of the existing commercial applications are discussed below.

CROSS-LINKED POLYETHYLENE

Among cross-linked materials produced, the most important by far are polyethylene wire insulation, shrinkable tubing, and plastic film.

Irradiated polyethylene, under the trade name Irrathene, was introduced as early as 1954 as tape and film for electrical insulation by the General Electric Company. Several companies now active in the production of wire insulation and shrinkable tubing include the General Electric Company, the Electronized Chemicals Corp., the Raychem Corporation, Radiation Materials, Inc., the Sequoia Division of Anaconda Wire & Cable Co., Suprenant, and Radiation Dynamics, Inc. Irradiated polyethylene wire insulation is more heat resistant than untreated polyethylene and has better stress characteristics. Conventional polyethylene film melts below 125°C, whereas the irradiated material maintains its integrity up to about 250°C. In addition, the irradiated film is fivefold stronger and can be stretched to twice its original size and then control-shrunk for contour-fit packaging.[4] The Cryovac Division of W. R. Grace & Co.[5] is the major producer of polyethylene food wrap today. Generally, an optimum balance of properties is obtained by a dose of 15 to 20 megarads. At present, machine sources of electrons are used exclusively for the processing of polyethylene products because of the minimal penetration required.

SEMICONDUCTOR COMPONENTS

Electronized Chemicals Corp., a subsidiary of High Voltage Engineering Corp., is processing well over a million semiconductors per month for a number of manufacturers of solid-state devices.[5] In this use radiation produces a controlled number of imperfections in a lattice by positive-ion proton implantations. These imperfections reduce the switching time of the device. Machine-irradiation costs for semiconductor applications vary between 0.5 and 5 cents per unit and often represent less than 1% of the selling price of the unit.

ETHYL BROMIDE

The radiation-catalyzed production of ethyl bromide by the Dow Chemical Co. was the first commercial radiation process for chemical production. Ethyl bromide, a volatile organic liquid used primarily as an intermediate for organic synthesis, is used to make pharmaceutical products.

The conventional aqueous process of reacting ethanol and hydrogen bromide yields water as a by-product.[6] The outstanding advantage of the

new process is that it produces a material virtually 100% pure without side products or contaminants. The reaction mechanism has been postulated[7] as follows:

$$Br^{\cdot} + CH_2=CH_2 \rightarrow BrCH_2-\dot{C}H_2$$

$$BrCH_2-\dot{C}H_2 + HBr \rightarrow BrCH_2CH_3 + Br^{\cdot}$$

Simple and reliable operation with efficient use of gamma rays from a 2000-curie ^{60}Co source makes the new Dow process more economical than that for conventionally produced ethyl bromide. In the new process ethylene gas and liquid hydrogen bromide are fed to the bottom of a reaction vessel and flow upward around a core of ^{60}Co. As the radiation-catalyzed reaction takes place, ethylene goes into solution as ethyl bromide.[8] G values in excess of 20 thousand are obtained. This process is the result of 7 years of research and development by Dow.[9]

STERILIZATION OF MEDICAL SUPPLIES

Surgical sutures can be sterilized by irradiation of the finished packaged product, a process that eliminates both heat and aseptic handling procedures. Compared to a tensile-strength loss of 12 to 22% from heat sterilization, a loss of only 4 to 8% results from radiation processing. Thus irradiated sutures are 8 to 14% stronger than heat-sterilized ones.[10] In addition, pliability is maintained. Of the sutures sold in the United States today, approximately 80% are sterilized by radiation. Ethicon, Inc., a subsidiary of Johnson & Johnson, is a large-volume producer of sterilized sutures although such companies as Hospital Supply Co., Inc., and Becton-Dickinson & Co. also irradiate medical supplies. Radiation sterilization of disposable syringes and needles is also increasing.

Until recently machine radiation was used exclusively as the radiation source; however, a reevaluation of technical aspects led Ethicon, Inc., to construct a plant in Texas that will initially use 60,000 curies of ^{60}Co. Table 198-1 gives the volume of commercial processes discussed above.

Process Applications

PROSPECTIVE

Commercialized processes represent initial returns on years of research investment. We expect that, with commercialization of the following processes, which represent only a few of the total potential applications under investigation, more substantial use of radiation will be realized in the near future.

Table 198-1 VOLUME OF EXISTING COMMERCIAL APPLICATIONS

Process	Annual volume
Cross-linked polyethylene wire insulation, shrinkable film	$27 million
Cross-linked polyethylene film	1500 tons
Semiconductors	20 million units
Ethyl bromide	400 tons
U. S. medical supply sterilization	$28 million

WOOD-PLASTIC MATERIAL

A new family of wood-plastic materials has been developed at West Virginia University under contract to the AEC.[11,12] The product, a wood-plastic combination, is produced by impregnating wood with a liquid monomer, such as methyl methacrylate, and then inducing polymerization by radiation. Various combinations of polymer and wood have been evaluated, including sugar maple, white pine, birch, and white oak. In general, the resulting product:

- Can be made fire resistant
- Is up to 900% harder than natural wood and hence is more resistant to blows, scratches, or marring
- Has a considerably higher compression strength
- Has much improved static bending strength
- Absorbs moisture more slowly and therefore has more dimensional stability (resistance to warping and swelling)
- Highlights the natural-wood grain and color
- Can be sawed, drilled, turned, and sanded to a hard, beautiful, satin-smooth finish

Probable markets include those for end-item or finished products. Typical applications include:

Furniture (indoor and outdoor)	Decorative trim
Floors	Structural beams
Window frames, sills, and doors	Sporting goods
Tool handles	Boat decks and fittings
	Shoe lasts
	Dies and jigs

Radiation doses required to induce the polymerization have been reduced by as much as

75% (to 0.5 megarad) in development work. Gamma energy from ^{60}Co is being used because of its deep penetration and independence of target shape. The processing of panels or veneers, however, may be better adapted to machine irradiation.

FOOD PROCESSING

Two programs within the United States relate to food preservation.[13–16] The U. S. Army Quartermaster Corps aims to develop selected radiation-sterilized (high dose) meat items for military rations that can be kept for long periods of time without refrigeration. The second part of the overall national program, an extension of earlier Army work, has been supported by the AEC since 1960. Its prime objective is the development of radiation-pasteurized (low dose) fish and fruit products. Fruit decay is reduced up to 75%, and the shelf-life of fresh fish is extended up to five times normal. A further objective is the development of prototype commercial food-irradiation facilities.

The Army's research is centered at laboratories in Natick, Mass. Their Radiation Laboratory includes a complete food-preparation area, a 1.1 million curie ^{60}Co source, and a 24-Mev 18-kw linear accelerator. Sterilization of ham, pork, chicken, and beef is being emphasized. Large-scale organoleptic tests performed during the past year indicate that all these items are now acceptable for rations.

Research in the AEC pasteurization program is being conducted at a number of universities and other laboratories throughout the country. Products showing the best potential are peaches, nectarines, oranges, strawberries, tomatoes, soft-shell clams, haddock, halibut, crab, flounder, and shrimp. Because of the lower doses involved in radiation pasteurization—generally less than 0.5 megarad—there are few, if any, changes in taste and texture. Significant advances in product development continue to be encouraging.[17–21] Several highlights of the overall national program are noteworthy:

On Feb. 8, 1963, radiation-sterilized bacon (dose, 4.5 to 5.6 megarads) was approved for unlimited public consumption. Acceptable radiation sources include ^{60}Co, ^{137}Cs, and electrons with energy of 5 Mev or less.

On Aug. 21, 1963, radiation disinfestation of wheat and wheat products was approved. Acceptable sources are ^{60}Co and ^{137}Cs. Approval of irradiation with 5-Mev electrons has been requested.

Petitions for sprout inhibition of potatoes (5000 to 10,000 rads) and mold and decay control of oranges (75,000 to 200,000 rads) are currently being evaluated by the Food and Drug Administration.

During the next 12 to 18 months, clearances will be requested for pasteurized fruits, such as apples, pears, sweet cherries, peaches, and nectarines; tentatively for marine products, such as halibut, haddock and related bottom species, and shrimp; and for chicken and pork. Clearance to irradiate onions for sprout control is also likely.

Radiation facilities for food processing either in operation or under construction[22] include those for bulk or packaged grain products,[23] an experimental on-ship irradiator,[24] a mobile truck-mounted irradiator,[25] and a land-based fish-processing pilot plant.[26] All use ^{60}Co.

Marketing and economic studies on radiation-pasteurized fish and fruit products indicate a price range acceptable to industry.[27] Nearly 60% of the U. S. fishing industry could tolerate a radiation cost of 1 to 3 cents per pound, which can now be met, and fruit-irradiation costs, neglecting savings in spoilage, generally should be 1 cent or less per pound.

CURING OF COATINGS

A radiation process developed by High Voltage Engineering Corp. of Burlington, Mass., has proved excellent for the curing of paint primers and coatings in building materials. Information is limited owing to proprietary interest. It is evident, however, that costs for production-line curing of factory-applied coatings compare favorably with those for conventional air-drying or heat-curing. The radiation-curing costs range from 30 to 80 cents per thousand square feet. Realistic production levels using machine-produced electron radiation are approximately 18,000 sq ft/hr as against an industrial average of about 12,000 sq ft/hr for conventional curing.

IMPROVED SEMICONDUCTOR DEVICES

Fundamental Methods Associates of New York is developing a technique known as "transmutation doping," which produces unique semiconductor components.[28] The semiconductor material is transmuted by thermal neutrons from a reactor to produce electronically active "dopant" impurities at desired spatial locations within the semiconductor crystal. Deposition of the dopant impurities is controlled by completely enclosing the semiconductor within a

"radiation die." This die is a thin boxlike structure composed of thermal-neutron absorbing material (e.g., boron or cadmium) with a pattern of slits. Neutrons enter the semiconductor in a geometry determined by the slit pattern. Diodes, transistors, microelectronic circuits, solar cells, and thermoelectric elements are typical products. The "proof-of-principle" of this process has been firmly established, and refined techniques are now being developed for the production of complex microelectronic units.

BIODEGRADABLE DETERGENTS

Esso Research and Engineering Company has developed a process for the radiation synthesis of a new sodium alkane sulfonate biodegradable detergent. The synthetic detergent material most commonly used is tetrapropyl-benzene sulfonate, derived from petroleum. More than one billion pounds is produced yearly. In the United States national and state legislation banning synthetic detergents that are not biodegradable has been introduced. In Germany a law to this effect has already been enacted. Because of such laws detergents must be developed which are not only biodegradable but which are also economical and have high cleansing characteristics. According to C. L. Fleming of Esso, their new detergent, the product of a radiation-induced sulfonation action, is made from an inexpensive common hydrocarbon fraction.[29] This new detergent is now being produced in ^{60}Co pilot-plant facilities by Esso.

TEXTILES

American Cyanamid Co. of Stamford, Conn., uses radiation to improve the dyeability of polypropylene. The process consists of radiation grafting of chloromethylstyrene to polypropylene textile fibers. Such grafted fibers can be dyed by direct or acid dyes in aqueous baths using levels of graft of about 25 wt.%. This leads to covalent bonding, arising from the reaction between the dye and the benzyl chloride group of the graft. Dyeing in standard aqueous baths can also be achieved at even lower levels of graft (less than 10 wt.%) if the grafted material is treated with a nucleophilic reagent such as pyridine. This improvement in the dyeability of polypropylene greatly widens its potential application as a fiber. American Cyanamid claims that with this radiation process polypropylene can be made as dyeable as wool at a cost of 5 to 10 cents per pound.

POLYMERIZATION OF ETHYLENE AND COPOLYMERS

Intensive research has been done at Brookhaven National Laboratory for the past few years on the radiation polymerization of ethylene and copolymers. This radiation-induced reaction proceeds with a high degree of efficiency (G values up to 150,000). The polyethylene produced is of high molecular weight. At the doses used the material is noncross-linked. Even more important, many copolymers of ethylene have been polymerized by radiation, the most important of which is a copolymer of ethylene and carbon monoxide.

High-density polymers cannot be produced easily in conventional processes; therefore copolymers starting with inexpensive raw materials such as ethylene and carbon monoxide are of great interest. The construction of a pilot plant for the production of these materials in quantities sufficient for industrial evaluation is planned in the near future.

Conclusion

Although the process radiation industry is still in its infancy, several processes now are a commercial reality, and others are close to it. Industrial firms as well as the U. S. Government are doing research on the use of process radiation energy. In conclusion two significant aspects should be noted:

1. Problems in the technological development of radiation processing are basic in nature and very wide in scope. Thus they are not specific to the particular product interests of any one industry or organization.
2. The research equipment and facilities needed to develop the required basic technology are novel in design, significant in cost, relatively expensive to operate, and often beyond the industrial capability of even specially trained and experienced personnel.

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The Development of Chemonuclear Processes

By B. Manowitz,* M. Steinberg,* J. W. Sutherland,* P. Harteck,† S. Dondes,† and J. H. Cusack‡

Paper 292 (abstracted)

In general, the research effort in the United States in chemonuclear processes is in an early stage of development. The major effort is being applied at three laboratories: Brookhaven National Laboratory (BNL), Rensselaer Polytechnic Institute (RPI), and Aerojet-General Nucleonics (AGN), with additional work being carried out at other universities and industrial laboratories.

The work at BNL and RPI is of a research and development nature; the chemistry and technology of a variety of chemical systems that lead to chemonuclear processes are being studied. The primary objective of the AGN program is to develop a pilot plant that will demonstrate the feasibility of producing hydrazine from ammonia by exposure to fission-fragment recoil energy.

It appears from economic data[1] analyzed at BNL that G values in excess of 1 are required. Single-purpose plants may have economic potential for the production of certain chemicals, e.g., hydrazine, ozone, and hydrogen peroxide, but a larger dual-purpose plant may be required for the fixation of nitrogen to produce fertilizer.

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†Rensselaer Polytechnic Institute, Troy, N. Y.
‡Aerojet-General Nucleonics, San Ramon, Calif.

Use of Power Reactors as Large-Scale Radiation Sources for Radiation Chemical Production

By F. Herre, W. Herzog, S. Rösinger, W. Schütze, and A. Wacker*

Paper 542 (extracted)

Although numerous radiation chemical reactions having high G values (such as polymerizations, chlorinations, sulfochlorinations, and addition reactions with hydrogen halides) have been investigated on a laboratory scale, only the manufacture of ethyl bromide from ethylene and hydrogen bromide has reached industrial proportions.[1] Because of a G value of 4×10^4 and the relatively low production rate of ethyl bromide envisaged, a ^{60}Co source of 2 kilocuries is adequate in this process. In general, considerably larger and more powerful radiation sources will be required in the future for other processes.[2–6]

Experiments have been done on the direct synthesis of nitrogen oxides by irradiation with fission products.[7–9] One of the difficulties of the process lies in the very short range of the fission products in uranium. Because of the short range, complicated fuel elements are required.

It is, however, advantageous to irradiate liquids with gamma radiation since a uniform release of energy is ensured even in larger reaction volumes. Three reactor configurations are feasible: the chemonuclear reactor,[6] the "blanket" reactor,[6] and the type suggested here—the "compound" reactor (Fig. 542-1).

In the chemonuclear reactor the reactants flow through the reactor core. In the blanket system the reaction space is arranged in a zone that is normally occupied by the reflector, i.e., within the reactor pressure vessel. In the compound reactor the chemical-reaction space is situated outside the pressure vessel, i.e., between the wall and the biological shield. In contrast to the chemonuclear and blanket reactors, the compound reactor requires negligible modifications of existing reactors.

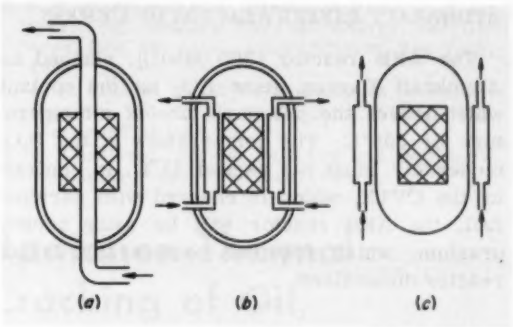

Fig. 542-1 Types of reactors for radiation chemical reactions. (a) Chemonuclear reactor, (b) blanket reactor, (c) compound reactor.

Nuclear Reactors Suitable as Compound Reactors

The chief requirement of a compound reactor is a high gamma-flux and a low neutron-flux density outside the nuclear reactor vessel. High thermal power alone is not sufficient. A high gamma-flux density exists inside the core of the pressurized-water reactor and the boiling-water reactor, but the thick walls of the reactor vessels will reduce the flux by several orders of magnitude so that they cannot be used for our purposes. The low gamma-flux density inside Calder Hall type reactors also precludes their being considered. Only reactors having a high power density and a pressure vessel whose wall thickness is relatively thin because of low operating pressures are of advantage. Such types are, for instance, pressure-tube reactors, (probably) sodium-cooled reactors, and organic-moderated reactors. A few pressure-tube reactors are discussed in detail.

CAROLINAS VIRGINIA TUBE REACTOR OF WESTINGHOUSE ELECTRIC CORPORATION[10]

The CVTR is a 60-Mw(t) D_2O reactor. Its construction enables a temperature of 68°C to be maintained in the reactor vessel. The coolant temperature in the tubes is nearly 300°C, and the pressure is 100 atm. Since the pressure in the moderator is about 1 atm gage, the wall of the moderator vessel can be kept thin (2 cm of aluminum).

*Farbwerke Hoechst AG, vormals Meister Lucius & Brüning, Frankfurt (Main)-Höchst, Germany.

ATOMKRAFT BAYERN REACTOR OF SIEMENS

The AKB reactor [300 Mw(t)], planned for Atomkraft Bayern, uses CO_2 as the coolant, which leaves the pressure tube at a temperature of 550°C. The temperature of the D_2O-moderator must not exceed 85°C. In contrast to the CVTR, which is charged with enriched fuel, the AKB reactor will be using natural uranium, which requires considerably larger reactor dimensions.

630A REACTOR OF GENERAL ELECTRIC[11]

The 630A reactor is air cooled, light water moderated, and highly enriched. Having a capacity of about 85 Mw(t), this plant delivers 90 tons of superheated steam per hour at 520°C and 60 atm. This high-steam-energy state has not been reached by any reactor so far. The high enrichment yields a great power density in the small core (height, 1 m; diameter, 1.8 m) and thus a high gamma-flux density outside the reactor vessel.

REACTOR MODIFICATIONS

Reactors are normally fitted with thermal shields that reduce the neutron and gamma radiation to a degree tolerable for the vessels used. For compound reactors these absorbers must be replaced by materials that preponderantly absorb neutrons. Materials in question are graphite, beryllium, and boron and their compounds and mixtures with water. Because of the favorable ratio of gamma absorption cross-section to removal cross-section, these materials reduce the gamma radiation only to a minor extent. A calculation carried out over six energy groups of neutrons for the General Electric 630A resulted in curves representing neutron-flux density in the chemical reaction space. It is possible to obtain gamma dose rates of several megarads per hour. If the thermal shield is replaced by a weak-gamma absorber, special precautions must be taken because of excessive gamma heating of the container materials.

ESTIMATION OF PRODUCIBLE AMOUNTS

The following data are required for the calculation of radiation chemical products: gamma dose rate outside the nuclear reactor vessel, volume and type of chemical reaction vessel, and G value of the corresponding reaction. The G values of several radiation chemical reactions of technical interest which are initiated by gamma radiation are compiled in Table 542-1. The dose rate at which each G value was measured is also shown since G values may depend on the dose rate. If several simultaneous reactions are considered, this could be effected by subdividing the chemical-reaction space, e.g., radially. The dose-rate dependence of low-G-value reactions is generally a linear function, whereas the dose-rate dependence of reactions involving chain reactions (high G value) is essentially a root function.

Table 542-1 CHARACTERISTICS OF RADIATION CHEMICAL REACTIONS

Reaction	Ref.	Molecular weight	G value, molecules per 100 ev absorbed energy	Dose rate, Mrad/hr
Hexadecane sulfonic acid from n-$C_{16}H_{34}$, SO_2, O_2	12	306	1260	0.09
Gammexane* from C_6H_6, Cl_2	13	288	3×10^5	0.001 to 0.01
Ethyl bromide from HBr, C_2H_4	1	109	4×10^4	0.25
Polymerizations of various monomers	14	50	10^3 to 10^6	

*Noted in the original manuscript as "Gammexane(R)"

The results available so far[12–14] permit estimations to be made of production quantities that can be attained using as the radiation source the power reactors discussed earlier (Fig. 542-2). An annular reaction space 1 m thick and equal in height to the core was assumed in order to compare the three reactors. This space, when filled with water, absorbs more than 90% of the radiation energy. It appears that only radiation chemical reactions

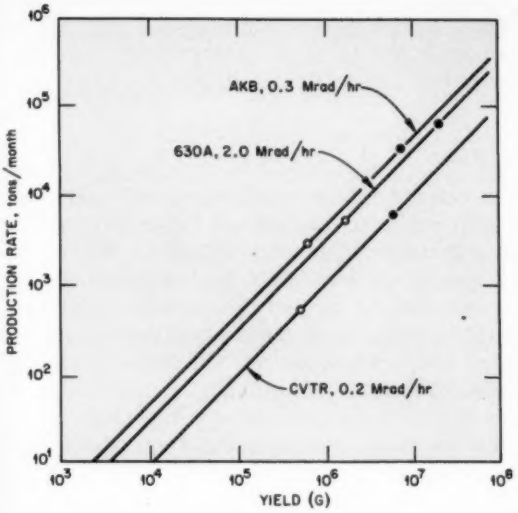

Fig. 542-2 Estimated production rates of hexadecane sulfonic acid (○) and ethyl bromide (●) in power reactors AKB, 630A, and CVTR.

having G values greater than 100 will be suitable for production on a technical scale in compound reactors. The quantities attainable for $G < 100$ are too low; if $G > 10^6$, a ^{60}Co source should generally be adequate.

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Radiation-Thermal Cracking of Oil, Oil Products, and Gaseous Hydrocarbons

By L. S. Polack*

Paper 389 (extracted)

Radiation-thermal cracking (RTC) of hydrocarbons can be accomplished by both radiation and heat.[1-8]

This paper reviews our investigations of radiation-thermal cracking under various experimental conditions of radiation using different hydrocarbon and oil stocks. The process was carried out in flow units using 0.8-Mev fast electrons from a linear accelerator and mixed gamma and neutron irradiation from a nuclear reactor. A study has been made of the effect of temperature, pressure, and dose rate on RTC product yields.

Fast-Electron Irradiation

RADIATION-THERMAL CRACKING WITH NONUNIFORM TEMPERATURE PROFILE[6]

Material was irradiated with 0.8-Mev electrons. The dose rate by acetylene dosimetry was 2×10^{16} ev/(sec)(ml) of vapors under normal conditions. The unsaturate content was determined by electrometric titration.[9]

n-*Heptane*. Experiments on the RTC of *n*-heptane, as with other feed stocks, were made at temperatures of 400 to 600°C. Radiation-thermal cracking occurs at much lower temperatures than thermal cracking (TC). Figure 389-1 shows that the unsaturate percentage in RTC rises very steeply with increasing temperature, exceeding 80%, while in TC it is only 50 to 55%.

*Union of Soviet Socialist Republics.

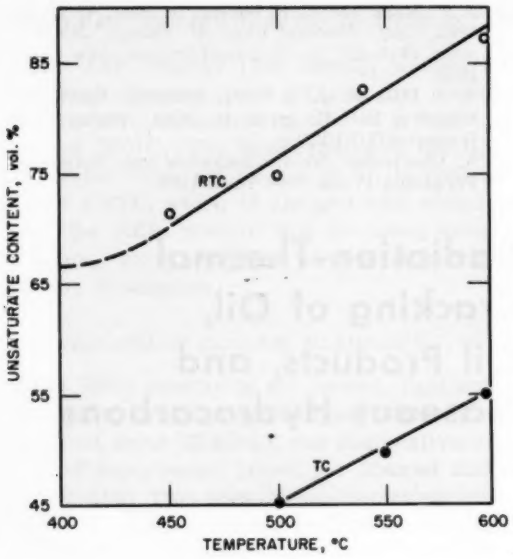

Fig. 389-1 Effect of temperature on unsaturate content of the C_2 to C_4 fraction for RTC and TC. (Editors' Note: These data have been replotted from original.)

140°C End Point (E.P.) Gasoline and Gas Naphtha. The gaseous product yield–temperature curves for RTC of these gasolines are similar in shape to those for *n*-heptane. The RTC gaseous products of 140°C E.P. gasoline are richer in olefins than those resulting from TC. In the RTC of heavy gasoline (200°C E.P.), the yields of gaseous products, including olefins, also increase with increased temperature.

Yields of gaseous products and the unsaturate distribution at the temperatures that were maximum in our experiments are given in Table 389-1. In the RTC of "middle-gas oil," an increase in yields of gaseous products is similar to that for the RTC of light feed stocks.

RADIATION-THERMAL CRACKING WITH UNIFORM TEMPERATURE PROFILE[10]

A reactor of new design was used to estimate feed conversions and yields of gaseous products as a function of process temperature. As can be seen from Fig. 389-2, the conversion in the experiments using the new reactor is almost twice as high as in the reactor with a nonuniform temperature profile. The yield of gaseous hydrocarbons, including olefins, much exceeds that in earlier experiments with another reactor. The flow unit used in these experiments is similar to that described in Ref. 11.

Radiation in a Nuclear Reactor

A special flow unit was developed for RTC in the channel of a nuclear reactor.[12] The unit was designed for a horizontal channel and can be used for flow processes at temperatures of 400 to 600°C and at pressures of 1 to 30 atm, the pressure being determined by the vapors of the feed itself.

Temperature–yield curves for gaseous products of TC and RTC at atmospheric pressure, as shown in Fig. 389-3, are similar in shape to those obtained in an electron accelerator[6,10] or from a cobalt source of radiation.[13] Table 389-2 shows the distribution of gaseous products of RTC of a 200°C E.P. fraction of condensate by mixed gamma-neutron radiation.

A comparison of the data given in Tables 389-1 and 389-2 shows that the gaseous-product

Table 389-1 REPRESENTATIVE DATA ON YIELDS OF GASEOUS PRODUCTS IN TC AND RTC

Feed	Temp., °C	Process	Yield, cubic centimeters per gram of feed								
			Total	H_2	CH_4	C_2H_6	C_2H_4	C_3H_8	C_3H_6	C_4H_{10}	C_4H_8
140°C E.P. gasoline	400	RTC	28.7	2.8	10.1	2.8	3.0	0.7	2.1	2.5	0.9
	600		206.0	15	58	16.9	43.2	3.9	26.2	15.1	13.1
	400	TC	–	–	–	–	–	–	–	–	–
	600		97	7.2	25.6	8.0	21.6	1.6	1.2	7.2	4.8
200°C E.P. gasoline	550	RTC	*	6.7	30.0	8.5	20.0	3.6	10.9	2.4	6.8
		TC	*	2.0	9.8	2.7	6.0	0.6	2.7	0.9	1.1
200°C E.P. fraction from crude	550	RTC	*	9.5	30.6	13.0	36.6.	1.2	13.0	2.4	9.5
		TC	*	2.2	8.7	3.9	9.1	0.2	3.2	1.5	1.9
250 to 363°C middle-gas oil	450	RTC	22.6	5.7	3.9	1.0	8.5	1.0	3.6	0.6	1.0
	550		100	3.0	19.9	13.5	33.6	4.0	16.6	–	9.9
	450	TC	2.7	0.34	0.81	0.3	0.85	0.05	0.17	0.07	0.05
	550		43	1.9	13.1	5.8	15.6	1.5	4.9	–	2.3

*Blank on original.

distribution is not appreciably affected by the mixed radiation. Table 389-1 shows that the unsaturate content of the gaseous mixture decreases with increasing temperature; however, the absolute yield of unsaturated hydrocarbons increases.

When the RTC results were compared with those of pyrolysis using the same feed stock, it was found that the total yield of RTC gaseous products at 570°C was 1.2 times as high as in pyrolysis at 77°C and the total unsaturate content was 1.5 times as high. With increasing temperature a considerable change is observed in the distribution of RTC liquid products.

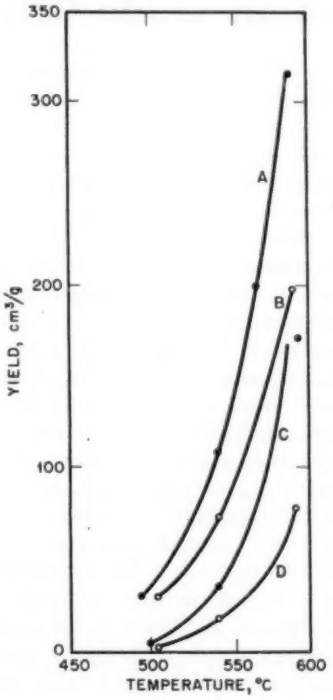

Fig. 389-2 Temperature conversion for 140°C E.P. straight-run gasoline. (A) RTC and (B) TC in reactor with uniform temperature profile; (C) RTC and (D) TC in reactor with nonuniform temperature profile.

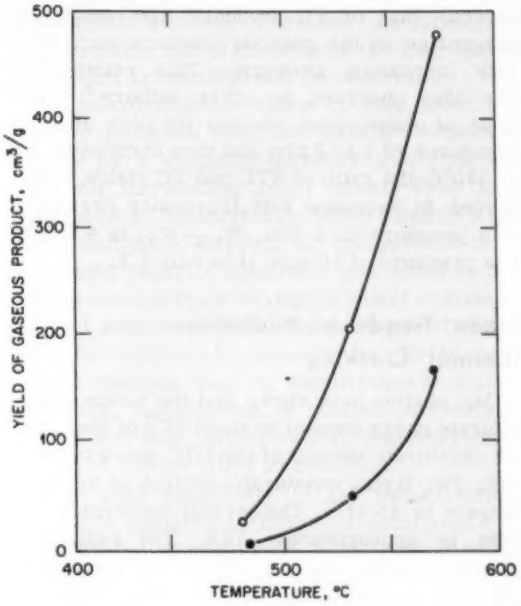

Fig. 389-3 Effect of temperature on gaseous-product yields for RTC(○) and TC(●) of the 200°C E.P. gasoline fraction at a pressure of 1 atm.

A kinetic treatment of the results gives a value of 60 ± 2 kcal/mole for the total activation energy for pure TC, which is in agreement with the reported value. The activation energy of the assumed pure radiation process defined by the difference between the rates of RTC and TC is 23 ± 3 kcal/mole. The latter value is in good agreement with the values obtained using other radiation sources and other feed stocks (Refs. 5, 6, 11, and 13). An assumed radiation-chemical yield of the RTC products estimated by the difference between the RTC and TC rates in terms of molecules per 100 ev of energy absorbed is 10^4 to 10^5 molecules per 100 ev, depending on temperature.

The total yield of gaseous products increases with increased pressure, linearly in the range of 1 to 10 atm, and the yield of RTC products

Table 389-2 GASEOUS-PRODUCT DISTRIBUTION FOR RTC AT DIFFERENT TEMPERATURES

Temp., °C	Distribution of gaseous products, %								
	H_2	CH_4	C_2H_6	C_2H_4	C_3H_8	C_3H_6	C_4H_{10}	C_4H_8	C_5H_{12}
480	11.2	42.1	13.4	25.2	2.8	–	5.3	–	–
530	7.4	43.5	10.6	20.2	1.6	12.2	2.2	0.9	1.3
570	6.6	54.7	6.5	19.4	0.7	8.6	4.3	0.6	1.2

exceeds that of TC products. The unsaturate percentage of the gaseous products decreases with increasing pressure. This relationship was also observed by other authors.[8,13] The yield of unsaturates reaches its peak at mean pressures of 1 to 3 atm and then decreases. Up to 530°C the ratio of RTC and TC yields is observed to decrease with increasing pressure; at a pressure of 1 atm, W_{RTC}/W_{TC} is 4.5, but, at a pressure of 10 atm, it is only 1.2.

Future Trends in Radiation-Thermal Cracking

Our studies have shown that the yields of unsaturate gases amount to about 55% of feed, and the unsaturate content of the RTC gases is about 70%. The liquid unsaturate content of the condensate is 45 wt.%. The overall conversion per pass is approximately 75%. The radiation-chemical yield reaches 10^3 to 10^5 molecules per 100 ev, depending on dose rate, temperature, and pressure. Optimum yields and conversions in RTC of various feed stocks can be obtained at a temperature range of 400 to 600°C and at pressures of 1 to 3 atm, depending on dose rate. Yields can be increased further.

Preliminary estimates show that the RTC process is more economical than pyrolysis in a tube furnace.

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Understanding the Natural Evolution of Petroleum: Radiation Effects on Crude Oils

By Umberto Colombo, Ennio Denti, Giuseppe Sironi, and Edoardo Zimmer*

Paper 830 (abstracted)

Presented here are the results of irradiation experiments (done in a swimming pool reactor) on some Italian crude oils in the presence of different sedimentary rocks and water and in environmental conditions similar to those known for sedimentary basins. The total radiation doses used were comparable to those absorbed in geologic times by organic material associated with more or less radioactive shales. Considerable radiation effects on the structure, distribution of molecular weights, and viscosity of irradiated crudes were found. Along with the formation of complex condensed molecules, light liquid and gaseous products are formed. Worthy of note is the fact that irradiation in the presence of certain natural catalysts, particularly when water-wet, leads to the formation of much less hydrogen and fewer unsaturated molecules than are obtained when crude oils are irradiated without catalysts. In some instances this might explain why, despite the primary role that radiation is believed to have played in the natural evolution of petroleum, neither hydrogen nor unsaturated products are very abundant in sedimentary basins. In connection with these experiments, it was found that irradiation of crude oil and of its distillation fractions in the presence of suitable catalysts brings about interesting chemical modi-

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fications that improve the quality of the oil relative to its subsequent refining and chemical utilization.

Continuous Methods of Tracing Chemical Changes in Fluids Under Gamma Irradiation

By S. Minc, Z. P. Zagórski, and R. Broszkiewicz*.

Paper 112 (abstracted)

For optical investigation on gamma-irradiated systems, a special ^{60}Co source was constructed. A spectrophotometer was used as a monochromatic source of light, and standard elements of this apparatus were used to measure the amount of light coming through the sample. The amount of light received by the photocells is enlarged by the Cerenkov radiation emitted by the sample in the gamma-radiation field. The true extinction of the solution was determined with the aid of a graph. All measurements were performed in cells made from very pure quartz, which does not darken in the radiation field. The described setup is used not only for the measurement of absorption but also for the emission of light. It is also possible to distinguish between Cerenkov radiation and fluorescence and to investigate the long-lasting glow after irradiation and its spectrum.

In polarographic studies it was found that gamma radiation does not disturb the performance of a dropping-mercury electrode except for introducing minor effects from local increases of temperature, which slightly widen the confidence interval of measurements. A saturated calomel electrode was found to be a reference electrode having excellent resistance toward radiation. Its changes of potential may also be explained by temperature effects alone. These changes have no significance in polarographical applications. A number of reference electrodes (e.g., silver–silver chloride) turned out to be too sensitive to radiation.

The high sensitivity of the measurements makes it possible to follow medium stable intermediates of radiolysis. In the case of water, we can study hydrogen peroxide and, indirectly, peroxyorganic compounds occurring as intermediate species in chain oxidation reactions. Eventually we can also study post effects.

It was found that the change in the oxidation state of iron proceeds under the irradiation at the expense of organic compounds and oxygen present in the solution, an important implication for radiobiology. Although iron ions occur in blood in other complex compounds, it seems that the investigated systems may present a most simple in vitro model of chemical phenomena proceeding during irradiation of blood in vivo.

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Section

V

Isotopes and Radiation Technology

Source Development

Radioisotope sources are especially suited to difficult-to-radiograph specimens: in field inspections, where access is limited; in routine nondestructive testing, where geometric problems call for the use of small or irregularly placed sources; and in inside-out and 4π radiography.[1] Large beta sources have potential advantages over the more commonly used ^{60}Co and ^{137}Cs gamma sources in radiation processing, especially in the irradiation of thin films or sheets.[2] A single paper that touched on both these topics was given at Geneva. One part of the paper dealt with applications of liquid and gaseous radioisotopes to radiography and the other with a megarad beta source.

Two Radioactive Sources Used in an Unconventional Way

Paper 862

Radiography with Extended Sources of Radioactive Liquids or Gases

By T. Cless-Bernert and K. Duftschmid*

To obtain good radiographic (optical) resolution, every radiographer desires high specific activity in sources of minimal size. It seems rather surprising therefore that radiographs of suitable quality can be obtained with sources that are extended over the whole volume of the examined object.

By filling tubes with radioactive gases or liquids, we were able to get good pictures of weldings and corrosion marks in tubes of different material and wall thicknesses. We used nuclides emitting low-energy gamma rays. Because of the softness of radiation, slight variations of thickness in the absorbing material produce large differences in the transmission of radiation and therefore clear contrasts on the radiographic film. Compared to this effect, the loss of resolution due to the large extension of the source seems of little importance.

Defects in a welding of a 0.3-mm-thick aluminum tube were clearly visible in a radiograph taken with ^{166}Ho in the form of a solution of $Ho(NO_3)_3$ (Fig. 862-1). The same solution was used to examine a 4-mm steel tube. Another welding of the 4-mm tube was radiographed with ^{133}Xe. Similar results were obtained (Fig. 862-2).

To compare the quality of radiographs taken with liquids and gases with that achieved by conventional methods, we examined tubes with

Fig. 862-1 Radiograph of a weld of a 0.3-mm aluminum tube filled with ^{166}Ho solution.

Fig. 862-2 Radiograph of a 4-mm steel tube filled with ^{133}Xe

*Österreichische Studiengesellschaft für Atomenergie G.m.b.H., Vienna.

well-defined variations in wall thicknesses. An aluminum tube of 6-mm wall thickness was prepared with eight rings on the outside of the tube; each ring was 2 mm wide and gradually increased in thickness from 0.1 to 0.8 mm. All the rings were easily detected, even the 0.1-mm one, and the contours of the rings were clearly visible. To simulate conditions found in looking for corrosion marks, we carved grooves 0.1 to 0.8 mm in depth and 2 mm wide inside a steel tube of 3-mm wall thickness. Again the carvings were clearly detectable in spite of the fact that the radiographic resolution was poorer because of the larger distance between the source and the photographic film. Since for inspection of possibly corroded tubes the detection of even small holes in the tube wall is more important than a clear reproduction of the exact shape of these holes, the results obtained by this method are suitable for this purpose.

The development of the described method had been stimulated by the practical need to get information about the rate of corrosion in steam vessels in a sugar plant. In this type of factory, production goes on only during part of the year; hence machinery might possibly be attacked by corrosion during the period of shutdown.

The accumulation of small amounts of retained water in the downward bends of the tubing system inside the steam vessel would make this part of the system especially susceptible to corrosion. Examination of blocks of narrow bent tubes cannot be done by conventional nondestructive methods. Therefore the lower part of the tubes of the whole block is cut off and replaced every second or third year. A destructive study of cutoff ends showed that only 30 to 40% of the tubes really needed to be replaced; the rest of them were not affected by corrosion.

By inserting enough ^{166}Ho solution into each tube so that the suspect parts of U-shaped bends were filled with the radioactive liquid, we detected the corroded tubes rather easily by radiography (Fig. 862-3). The corrosion mark seen on the radiograph could easily be identified by deformations. The wall thickness of the tubes examined was 3 mm; the maximal depth of the defect shown was 0.8 mm.

The specific activity of the ^{166}Ho used was about 30 mc/liter and the exposure time was 15 hr. Because of the softness of radiation and the rather short half-life of ^{166}Ho (27.3 hr), the arrangements for radiation protection were relatively simple.

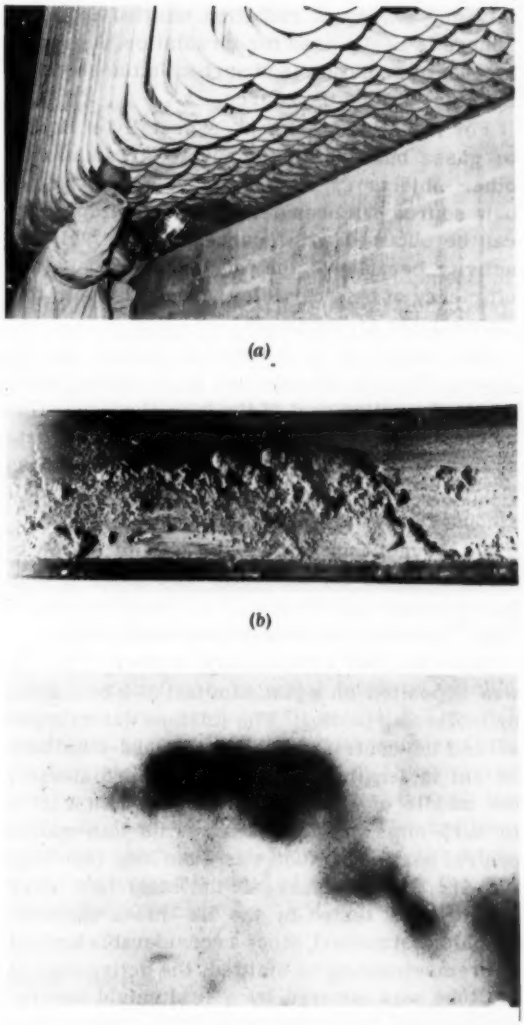

Fig. 862-3 Nondestructive inspection of blocks of narrow tubing of steam vessel of sugar plant. Photographs show (a) outside and (b) inside of one pipe that cannot be inspected by conventional nondestructive means. (c) Radiograph of pipe using ^{166}Ho solution.

A Megarad Beta Source

By T. Cless-Bernert and N. Getoff*

In conventional multicurie gamma-irradiation facilities, only a small percentage of the inten-

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sity and energy of radiation emitted from the source is really used for irradiation. A greater amount of radiation is absorbed in the shielding material around the source.

For radiation treatment, especially of liquids or gases but also for the surface irradiation of other objects of suitable size, a tube-shaped beta source has been developed. A high dose rate can be obtained with sources of relatively low activity because of the much higher ionization efficiency of beta particles, especially with this geometrical arrangement.

The liquid to be treated is poured into or passed through the tube and is irradiated while inside the active part of the tube. Contamination of the liquid is prevented by covering the beta source with a thin sheath of material whose weight per unit area is so low that only a small amount of the beta radiation is absorbed.

We have found that a high-quality stainless steel, such as the austenitic chromium-nickel-molybdenum-alloyed steel,* in combination with $^{90}Sr/^{90}Y$ meets the requirements sufficiently.

For the first experimental source, ^{90}Sr-^{90}Y was deposited on a platinum foil of 5 by 3.3 cm by molecular plating.[3] The foil then was wrapped around the central part of a stainless-steel tube 26 cm in length and about 11 mm in diameter, the middle of which was turned down on a lathe to 0.15-mm wall thickness. This thin-walled central part of the tube was 5 cm long (see Fig. 862-4). The tightness of the steel tube after turning was tested by the He–mass spectrographic test method. Since a considerable amount of bremsstrahlung is emitted, the active part of the tube was covered by a lead shield several centimeters thick. The cover of the source was rendered leakproof by means of two tightly fitting brass cones welded onto the steel.

Figure 862-4 shows the source, the lead cylinder, and the two leakproof locks. The ends of the steel lining protrude on both sides of the cylinder. The liquid is pumped into the source through polyvinylchloride (PVC) tubes mounted on the end of the steel tube.

Sources of this design give an average dose rate of 10^5 rads/hr. The dose rate per cubic centimeter in a liquid achieved with this arrangement depends on the amount of $^{90}Sr/^{90}Y$ that is deposited per square centimeter on the platinum foil. The molecular plating method is

*Stainless steel MASO, Schoeller-Bleckmann.

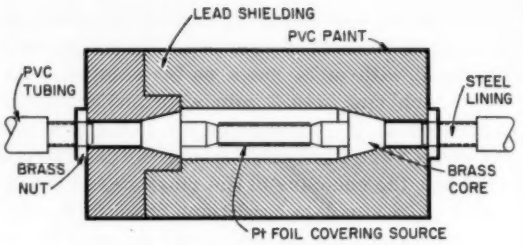

Fig. 862-4 Tube, beta source, and shielding.

an easy way to deposit thin layers of radioactive material of practically no self-absorption. The amount of the deposit, however, is limited to less than 1 mg/cm^2. With a strontium solution containing about 10 mg of solid material per curie, between 1 and 2 curies of $^{90}Sr/^{90}Y$ can be plated on the foil. Dose was measured with a Fricke dosimeter solution that was poured into a thin PVC bag having a wall thickness of a few milligrams per square centimeter. The bag was pushed into the active part of the steel tube.

The dose rate of the source can be increased by a factor of 2 if not only the foil wrapped around the steel tube is plated but also the steel tube itself. Safety is not compromised because of the high anticorrosive quality of the steel. For practical use a number of tube sources can be mounted in line, depending on the dose required. Since the diameter of the source cannot be increased much without reducing the dose rate within the source, several sources can be mounted in parallel if a large volume of liquid is to be treated.

Experiments are being done to find further ways of constructing a strong tube-shaped beta source of high energy, e.g., with tubes of flat rectangular cross-section instead of a circular one. Also, long-term tests are being conducted to find whether strontium diffuses through the steel, which seems very unlikely. If these tests prove to be satisfactory, this source would be a simple device for the irradiation of liquids and other materials not only in laboratories but also on a very large scale in industry.

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Section

VI

Isotopes and Radiation Technology

Isotopic Power Development

Isotopic power development discussions in previous issues of this Review have included safety aspects;[1] USAEC policies, requirements, and potential applications;[2] and a survey of radioisotope fuels development and operating power sources.[3] The two Geneva papers covered in this issue—one from the United States and one from Russia—summarize recent work on isotope thermoelectric converters. The U. S. paper is general and discusses the design of generators, the status of the technology, and areas for broader use. Hence it is presented in some length. The Russian contribution, on the other hand, reports in detail on two generators—their ^{210}Po and ^{144}Ce heat sources. Only an extract of the main points and representative data are given here. The reader is referred to the full paper for physical descriptions, results of tests, volt-ampere characteristics, temperature distribution, and activity of the device.

Workers in the field of isotopic power sources as well as those interested in direct conversion of nuclear heat into electricity have mutual interests in materials, methods, and theory, and cross-fertilization of ideas is possible. A number of papers presented at Geneva dealt with direct conversion of energy. An abstract of one of them is given here along with a list of the other papers.

Isotopic Power Sources Coupled with Thermoelectric Converters

By J. G. Morse and D. G. Harvey*

Paper 217

Exploitation of the principle of generation of electrical energy from the decay heat of radioisotopes has resulted in the development of compact and reliable direct-conversion power sources capable of long-term unattended operation. Feasibility of this technology was established under the USAEC Systems for Nuclear Auxiliary Power (SNAP) program which began in 1956 and led to the fabrication of a number of systems now providing dependable power for satellites in outer space, weather stations on land, and navigation buoys at sea.[4]

Design Considerations

A thermoelectric generator consists of a sealed capsule of radioisotope fuel enclosed in a heat accumulator or metal block on which a thermoelectric converter is mounted. Thermal insulation assures efficient heat transfer from the fuel capsule to the converter. Unconverted heat is rejected to the environment through the outer surface of the generator. The environment to which heat is rejected and the heat-transfer technique used depend upon the end use of the power plant.

Heat Source

Since the driving thermal energy results from the absorption of radiations from the encapsulated radioisotope, proper selection of the fuel and its containment is basic to the design of the device.[3] Half-life must be compatible with mission objectives; high power density (watts per square centimeter) is necessary to minimize size and weight; the type of radiations from the source and its daughters in secular equilibrium dictates shielding requirements; chemical form of fuel must satisfy safety requirements; and cost and availability should be consistent with the mission.

Fuels are obtained by purifying fission products or irradiating suitable target materials. Fissioning of uranium yields several hundred

*Small Power Systems Department, Martin Company.

radioisotopes. Of these, only four radionuclides are applicable to power generators (Table 217-1). Several predictions of quantities of fission wastes in a growing reactor economy have been made. One such estimate for ^{90}Sr suggests 1 billion curies [5 Mw(t)] by 1980. This estimate indicates that large quantities of fuel materials will be available for remote power applications.[5]

Irradiating suitable target materials yields alpha-emitting radioisotopes that, although more costly than fission products, are particularly important for certain missions, such as space flights,[6] where little or no radiation or shielding weight can be tolerated. Certain alpha emitters undergo spontaneous fission and also enter into alpha-neutron reactions when combined with elements of low atomic number.[6,7]

Table 217-1 PROPERTIES OF RADIOISOTOPES FOR HEAT SOURCES

Nuclide chemical form	Type of decay	Half-life	Specific activity, thermal			Availability, Mc/year*		
			Watts/g	Watts/cm³	Curies/watt	1964	1970	1980
$^{90}SrTiO_3$	β	27.7 years	0.21	0.7	154	3	25	100
^{137}Cs (glass)	β,γ	27 years	0.072	0.215	210	1	25	100
$^{147}Pm_2O_3$	β	2.6 years	0.18	1.0	2700	0.1	50	150
$^{144}CeO_2$	β,γ	285 days	2.3	13.8	128	1	200	4000
^{210}Po (metal)	α†	138 days	140	1320	31.6	0.1	3	30
$^{242}Cm_2O_3$	α	162 days	120	1170	27.6	0.06	0.3	
$^{244}Cm_2O_3$	α	18 years	2.5	22.5	28.6		0.2‡	
^{238}Pu (metal)	α	90 years	0.48	9.3	29.0	0.1	0.6	1.5

*AEC Isotopic Fuels Program, Division of Isotopes Development; presentation by W. K. Eister to Chicago Biology and Medicine meeting, Jan. 30, 1964.

†Gamma abundance is 0.013% at 0.8 Mev.

‡Only scheduled production, but quantities could be equivalent to ^{238}Pu after 1970.

Fuel Encapsulation

Large quantities of fuel are required for power generation; the release of this fuel could cause a significant hazard. Absolute containment of the fuel therefore must be assured until decay has rendered the isotope harmless. Space generators use multiwalled cobalt or nickel-base superalloys in cylindrical geometry with welded end closures. Such capsules have been extensively tested with simulated fuels and have been found to satisfy the stringent containment requirements for aborted missions: fire and explosion on the launch pad (3200°C for microseconds, falling off rapidly to 900 to 1600°C); blast overpressures (700 atm); and ascent abort (high-velocity impact against granite targets at operating temperatures). Testing has also assured containment of fuel when the capsule is subjected to high internal pressures. This test was necessary since alpha emitters, such as ^{238}Pu and ^{210}Po, release helium atoms upon decay.[8]

Terrestrial systems rely on Hastelloy C, a nickel-base alloy, for primary containment. Tests show that the rate of seawater corrosion of this material is less than 0.00025 cm per year. Containers can be designed with wall thicknesses that can withstand an ocean environment for more than 20 half-lives of ^{90}Sr.

Energy Conversion

The phenomenon of thermoelectricity was described first by Seebeck in 1832, who noticed that a temperature difference imposed across a junction of two dissimilar metals will generate an electric potential.[9] Intensive investigations to develop suitable semiconductor materials for this use have yielded Bi_2Te_3, PbSnTe, GeSi, and a few others, each with its attendant advantages and disadvantages. The conversion efficiency is directly related to Carnot efficiency and to the figure of merit, Z, a term that is dependent upon both properties of materials and temperature:[10]

$$Z = \frac{a^2}{\rho K} \tag{1}$$

where $Z = {}^\circ C^{-1}$

ρ = electrical resistivity, ohm-cm

a = Seebeck coefficient, volts/°C

K = thermal conductivity, watts/(°C)(cm)

The dimensionless number ZT is plotted in Figs. 217-1 and 217-2 for these materials in

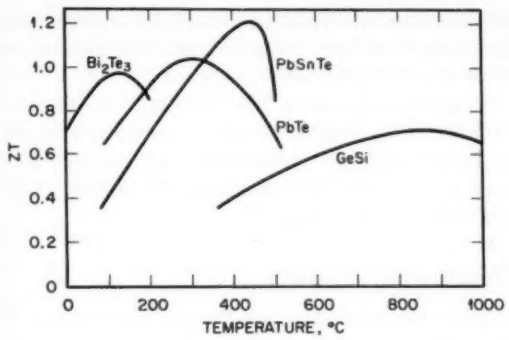

Fig. 217-1 Performance characteristics of N *materials.* ZT *is a dimensionless number (see text).*

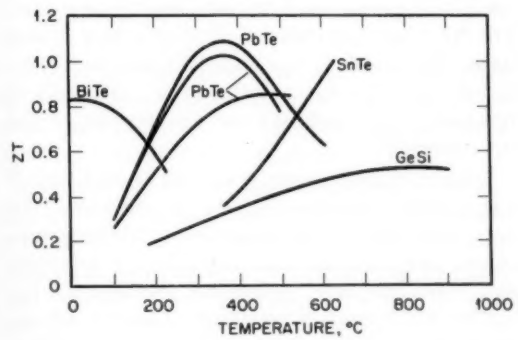

Fig. 217-2 Performance characteristics of P *materials.*

both N and P doping conditions. For maximum conversion efficiency, materials are selected for highest average values over the temperature range determined by the boundary conditions, namely, heat-source and radiator temperatures. If weight and size are to be reduced, space systems must have higher radiator temperatures than terrestrial systems. Higher power-to-weight ratios are obtained, however, at the expense of efficiency.[10]

Figure 217-3 illustrates the typical characteristics of a thermoelectric generator, all of which are predictable on the basis of simple theory. The generator behaves as a voltage source and, when connected in series with a resistor, yields maximum power at a point of matched impedance. The power curve for a given generator is obtained by first measuring variation of voltage with current from short to open circuit under conditions of steady-state performance. Peltier cooling will affect the hot-side temperature slightly. This temperature is at a maximum under conditions of zero current since none of the heat is converted into electricity; it is lowest at short circuit where maximum Peltier cooling occurs. The nonlinearity in the voltage vs. current curve is attributable to the variation of the property of the material with temperature. The internal-resistance curve also reflects this nonlinearity. Open-circuit as well as short-circuit voltage can be computed from the Seebeck voltage and the temperature range of operation. All other properties can be determined from temperature range and property data.

Examination of these characteristics reveals how the radioisotope thermoelectric generator can be used most effectively. The nature of the power curve indicates that the system is ideal for conditions of constant, continuous power. It will not, however, tolerate significant deviations in load voltage from the maximum power point. In addition, the device makes an excellent battery charger in that its voltage will follow that of the battery and will trickle charge without boiling off the electrolyte. Thus it is suited for furnishing power to systems requiring high peak power on a partial-duty cycle. Furthermore, it is a high-current low-voltage device requiring series (or series-parallel) connections within each bank of thermocouple pairs to increase the overall voltage. A static, solid-state, d-c-to-d-c converter is often used to match the electrical characteristics of the power supply to the requirements of the payload. This converter has operated at efficiencies of

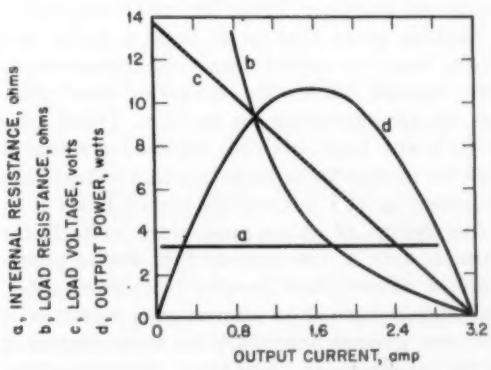

Fig. 217-3 Electrical characteristics of a thermoelectric generator.

up to 92% between 3 and 30 volts. The efficiency is largely dependent on the degree of regulation required.

Power Flattening

Unique with radioisotope power technology is the reduction of thermal input power with time as the fuel decays. With radioisotopes of short half-life, such as ^{210}Po and ^{242}Cm, a controllable thermal bypass is introduced around the thermoelectric elements to maintain a constant hot-side temperature over the mission life. In SNAP 11, a thermal shutter exposed the fuel capsule to space at the beginning of life by means of a thermal sensing device operating by the expansion of a liquid metal. The SNAP 7 series uses a thermally conductive gas that can be replaced by a less conductive gas after an interval of several years to raise the hot-side temperature to around its initial value. However, with generators fueled with long-lived isotopes, such as ^{238}Pu or ^{90}Sr, a small initial overload of inventory and operation below the maximum power point gives essentially constant power for a 10-year period.

Generator Safety

Generator safety is discussed in Ref. 11. Although reliance is placed first on fuel containment, generator systems are tested against environmental criteria related to their end use. These include thermal and mechanical shock and vibration peculiar to the mission. Terrestrial generators have been fueled with $^{90}SrTiO_3$, a chemical compound that has demonstrated extremely low solubility in freshwater and seawater,[12] good thermal conductivity, a melting point that is at least a factor of 2 higher than the center-line temperature of the fuel capsule within the operating generator, and no decomposition on melting. These generators are fully shielded; depleted uranium is used to attenuate the radiation flux to less than 10 mrads/hr at 1 m from the center line.

Any design of space generators reflects the ultimate fate of the nuclear fuel. Under conditions of uncontrolled reentry from earth orbit, fuel should burn up to micron-size particles at altitudes greater than 30.5 km to be dispersed in the stratosphere. However, because of extensive tests the orbit selected for SNAP 9A reflects a lifetime that is equal to about 10 half-lives of ^{238}Pu. Alpha emitters could conceivably be designed to reenter intact by recognizing the weight penalty required for the ablator or heat-shield materials.[13]

Launch trajectories are analyzed carefully to determine the region of impact in the much less probable event of partial burnup. Launch azimuths are selected with care to confine these regions to ocean areas.[11]

Operational Systems

With the launching of the modified SNAP 3 and SNAP 9A systems, the first use of nuclear power in space was achieved. The initial satellite was orbited in June 1961. Although the first units shared the power load with companion solar cells, after less than a year of operation the SNAP system was the only source of power. The first one continues to provide power at this time. The second, launched in November 1961, failed suddenly six months later. Analyses of telemetry data indicate the failure was not in the generator.

Specifications of space and terrestrial SNAP generators are given in Ref. 2. Projects IMP (Interplanetary Monitoring Probe), Nimbus, Surveyor, and ComSat (Communications Satellite) will use advanced generator technology for higher performance systems. ComSat anticipates using ^{90}Sr, which will require further safety development, particularly the ability to handle the fission product at the launching pad with minimum perturbations during the countdown procedures.

The first operational terrestrial system to use radioisotope power sensed and transmitted weather data at 3-hr intervals in an unattended remote arctic weather station for more than two years.[14] After exceeding its design life by several months, a failure in the electronics system required dismantling of the station. The generator is awaiting reinstallation this summer with new electronics equipment. In the SNAP 7 series, modularization of the thermoelectric elements has contributed markedly to the ease of assembly. Advanced systems will employ internally shielded thermoelement modules and more efficient thermal insulation; these changes will yield lighter, higher performance devices.

Advanced development is focused on improved thermoelectric materials as well as on more effective use of existing materials. The latter relates to Figs. 217-1 and 217-2, in which the

most effective use of the compositions noted is restricted to a narrow portion of the temperature range. At present thermoelectric configurations in operational systems are composed of P and N legs of a single composition. Further advantages will result when elements are constructed as composite materials that can operate over a wider temperature range. A temperature profile along the axis of such an element would indicate the transition of one material to another corresponding to the intersection of their ZT curves. The highest average ZT for each material would be obtained, and this would maximize the potential efficiency. Although values of about 20% are theoretically possible, practical considerations, such as heat sink, source temperature, and thermal leakage for a given application, tend to reduce this value.

Other factors, such as material-interface problems and compatibility, will likewise impose limitations; but the approach is nonetheless worthy of intensive investigation.

Further considerations for technological improvement will include efficient high-temperature thermal insulation and heat-source, cladding, and encapsulation materials, all of which will provide a basis for the designing of truly advanced generators.

Conclusions

Isotope power sources are compact and lightweight, have no moving parts, and are reliable for a long term. In space these systems are completely independent of the sun, and thus the need for reliance on vehicle placement, orientation, and complex solar mechanisms is eliminated. They are insensitive to radiation belts and lend themselves to long-lived satellites and to lunar and planetary probes. Their useful power range, which extends to the low-kilowatt region, offers a power capability to satisfy a number of manned-mission requirements.

Terrestrial systems have demonstrated continuous reliable performance, which recommends their use in inaccessible places for unattended applications. These have already included automatic weather stations in polar regions and at sea, navigation aids, and signal devices at the bottom of the ocean. A barge weather station using SNAP 7D has been developed. These applications will soon encompass networks of remote weather stations providing data to computation centers, radionavigation aids for aircraft, telephone-repeater power supplies, and precise undersea markers for shipping and transportation.

Radioisotope power systems have been developed with caution and responsibility with respect for the inherently hazardous nature of the materials. Each mission was judged safe only after extensive testing. This effort will continue for each system.

Although the cost of fuel materials is the dominant one, it is more often a relatively small factor in the overall mission cost, particularly in space, where long-term reliability and low weight are considered along with other ways of providing power. Terrestrial radioisotope units reduce considerably the logistics of resupply and will be truly competitive when (1) the equipment requiring power has both life and reliability compatible with those of the power source and (2) fission products become available in massive quantities from the now-planned USAEC production facility.[4]

Radioisotope-fueled thermoelectric generators have already shown, and continue to demonstrate, their utility and practicability to provide reliable long-term power for missions involving all regions of earth and outer space.

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Radioisotope-Fueled Thermoelectric Generators

By A. N. Voronin, I. G. Gverdtsiteli, R. S. Orinberg, Yu. D. Gubanov, V. A. Zharkov, V. F. Zhukov, V. K. Kovirsin, V. M. Kodyukov, N. P. Korotkov, R. Ya. Kucherov, S. P. Lalikin, A. I. Ragosinski, G. M. Fradkin, B. L. Shinderov, and S. Ya. Erman*

Paper 318 (extracted)

Radioisotope Power Source Using ^{210}Po

In recent years many papers have been published which describe various radioisotope-fueled thermoelectric generators delivering from 1 to 100 watts. Lead selenides and tellurides are used as a thermoelectric material because of their higher figure of merit, Z (see Paper 217), as compared with the well-known materials. The figure of merit, however, is not the only factor that indicates suitability of thermoelectric materials. Application of high-temperature alloys (although with less figure of merit) allows one to increase the emitter temperature and so to save weight while maintaining the efficiency as a result of the greater temperature difference between hot and cold junctions.

We designed a number of thermoelectric generators with initial power up to 10 watts. These had similar thermoelectric conversion systems and differed only in the radiator and in the way the thermopiles were pressed to the hot surface. A radioisotope heat-source simulator was used to determine equipment capacity, electrical parameters, and temperature distribution through the blocks.

In the simplest design a flat capsule in the shape of a rectangular parallelepiped 60 by 60 by 13 mm is snugly fitted between hot surfaces of two thermopiles, and the cold junctions of the thermocouples are attached to the radiator. If a capsule is to be reliable at pressures up to 200 atm (caused by helium produced in the decay), its surface temperature should not exceed 850°C. Temperature-resistant rubber used for sealing the device limits the temperature of the radiator, which should not be more than 250°C. The semiconductor section of the thermoelectric elements was 3 cm long. Silicon-germanium alloy used in this device has an average Z of 4×10^{-4} °C^{-1} and a thermal conductivity factor of 1.5×10^{-2} cal/(cm)(sec)(°C). The parameters of the device are as follows:

Insulation thermal resistance, (°C)(sec)/cal	12
Heat flow through insulation, watts	49
Initial heat power of the capsule, watts	320
Radiator surface, cm^2	900
Number of thermoelements (allowing for the external load voltage of 1.8 volts)	32
Thermopile cross-section, cm^2	0.8
Hot-junction temperature, °C	760 to 775
Cold-junction temperature, °C	300 to 330
Thermoelectric generator efficiency, %	3.5 to 3.9
Overall efficiency, %	3 to 3.3

Each battery of the generator comprised 16 thermoelements connected in series (8 P type and 8 N type).

The housing of one installation was of Duralumin and was cylindrical. The housings of two others were of Duralumin throughout and were more rigid and lighter.

The electrical simulator of the radioisotope heat source was a graphite unit 60 by 60 by 13 mm with a molybdenum hairspring. Heater power was determined by connecting an a-c wattmeter into the heater circuit. These systems were shown to be stable during 1000 hr of operation. Representative data are as follows:

Dimensions, mm	
Height	128
Diameter	195
Weight (without the radioisotope heat-source capsule), kg	3.1
Simulator heat power, Q, in watts	
Heat power equivalent of the radioisotope heat source containing 10,000 curies of ^{210}Po	320
Heat power equivalent after 3 months of operation	205
Electric power of the installation, watts	
When Q = 320 watts	10.15
When Q = 205 watts	4.35
Efficiency when Q = 320 watts, %	
Thermoelectric	3.75
Thermal	85
Overall	3.18
Electromotive force when Q = 320 watts, volts	3.9
Electromotive force when Q = 205 watts, volts	2.42
Maximum temp. of the thermoelements hot plate when Q = 320 watts, °C	861
Radiator maximum temp. when Q = 320 watts, °C	241
Maximum temp. of the thermoelement hot plate when Q = 205 watts, °C	670
Radiator maximum temp. when Q = 205 watts, °C	190

The efficiency and electric power of a thermogenerator with a nuclide heat-source simulator were also determined under various temperature conditions.

*Union of Soviet Socialist Republics.

The radioisotope heat source was a rectangular parallelepiped made of stainless steel. Capsules containing ^{210}Po were inserted in five cells. The nickel capsule containing 1000 to 2000 curies of ^{210}Po was mounted within two stainless-steel tubes for strength. Tubes were provided with screw stoppers that were welded during assembling. The source thermal power was determined calorimetrically.

In tests of the thermoelectric generator, a nuclide heat source containing ^{210}Po (the original thermal power of which equaled 244 watts) was set up for 50 min and then connected to a-c wattmeters. The following parameters were continuously controlled: the voltage produced by the thermogenerator across the 0.34-ohm load; the hot skin temperature (two points); the radiator housing temperature (two points); and the gamma-radiation dose rate at a distance of 0.3 m from the source. Some of the test results are listed in Table 318-1.

Table 318-1 REPRESENTATIVE RESULTS WITH ^{210}Po THERMOELECTRIC GENERATORS

Parameters	Operation period, hr			
	48	1000	1400	2000
Thermal power of the radioisotope heat source, watts	244	200	185	163
Electromotive force, volts	2.85	2.3	2.09	1.88
Load voltage at 0.34 ohm, volts	1.4	1.14	1.04	0.92
Power output, watts	5.8	3.79	3.24	2.52
Installation efficiency, %	2.36	1.91	1.75	1.55
Thermoelectric generator hot-plate temp. when operating at 0.34 ohm, °C	759	633	600	545
	746	542	503	440
Radiator housing temp., °C	230	202	192	163
	216	190	178	157
Activity of the radioisotope heat source using ^{210}Po, curies	7700	6300	5320	5140

Electric-Power Generating System Containing ^{144}Ce

The generator under consideration is a prototype of an electric-power source for automatic meteorological stations. The possibility of using short-lived beta-emitting nuclides in generators was determined by a ^{144}Ce test-model source during a controllable (constant output power) and an uncontrollable (output power falls off) period.

At the beginning of an uncontrollable period, the nuclide-fueled generator contained 17,500 curies of ^{144}Ce. During an uncontrollable operation the electric output was 5.0 to 5.6 watts; the voltage was 3.5 volts; and the overall efficiency was 3.5 to 4%. A thermopile based on solid solutions of $Bi : Bi_2Te_3 + Bi_2Se_3$ (N type conductivity) and $Bi_2Te_3 + Sb_2Te_3$ (P type conductivity) was used to convert the heat into electricity. A special automatic device provided a predetermined power level. The electric power was consumed by a storage battery with a capacity of 12 amp/hr. This battery supplied power to a weather station working in pulsed operation with 26 volts at 6 amp.

Radiation shielding consists of operating and shipping containers. The operating-container shielding was designed to provide a dose rate of less than 1.0 r/hr at 1 m from the generator surface. With the operating container inserted into the shipping container, the dose rate is reduced to less than 10 mr/hr at 1 m from the outer surface. The operating container has removable stiffening ribs, and the shipping container, fixed ribs. The generator may be used in the operating container alone or in both containers. Tungsten and lead are used as shielding materials. The total weight of the generator with the operating container is 400 kg; the shipping container weight is 1200 to 1400 kg.

An operating generator comprises a radioisotope (or heat) block, a thermopile, a thermal-power controller, an arrangement for substitution of the thermopile, housing, and biological shielding. [Editor's note: In the complete paper, these parts are described in some detail as are thermophysical and heat-generation calculations and shielding design.]

The thermoelectric conversion device consists of 97 thermoelements (5 by 5 mm and 20 mm high); the thermoelements are made up of solid solutions of the triple alloys $Sb_2Te_3 + Bi_2Te_3$ and $Bi_2Te_3 + Bi_2Se_3$. The generator tests were done with an electrical simulator of the heat source under normal operating conditions.

The ^{144}Ce generator described above is now being used as a power source at an automatic weather station located in the middle part of the Soviet Union. The generator is buried in the ground to minimize radiation.

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High-Temperature Direct-Conversion Reactor "Romashka"

By M. D. Millionshchikov, I. G. Gverdtsiteli, A. S. Abramov, L. V. Gorlov, Yu. D. Gubanov, A. A. Efremov, V. F. Zhukov, V. E. Ivanov, E. A. Koptelov, V. G. Kosovskii, N. E. Kukharkin, R. Ya. Kucherov, S. P. Lalikin, V. I. Merkin, Yu. A. Nechaev, B. S. Pozdnyakov, N. N. Ponomarev-Stepnoy, E. N. Samarin, V. Ya. Serov, V. A. Usov, V. G. Fadin, V. V. Yakovlev, M. V. Yakutovich, V. A. Khodakov, and G. V. Kompaniets†

Paper 873 (abstracted)

The "Romashka" experimental power plant uses a system in which heat generated in the reactor core is transferred by thermal conduction into a thermoelectric conversion device mounted on the external surface of the reflector. The application of beryllium as reflector, graphite as core, and uranium dicarbide as fuel has made possible the use of a high-temperature conversion device of silicon-germanium semiconductors. The conversion device is mounted on one radial and two end-face reflectors on the vertically positioned cylindrical reactor. Thermal insulation is used to reduce heat leakage at the reactor ends. Each thermoelectric pair is made up of N and P type thermoelectric elements connected by a switch plate on the hot side. On the cold side separate pairs are coupled into a single circuit. In all the device is divided into four groups of thermoelements, each of which has independent terminals. This design enables one to perform individual, series, and parallel experiments. Selected parameters of the device are:

Electrical output, kw	0.50 to 0.80
Total thermal power, kw	40
Maximum temp. of beryllium reflector, °C	1200
Maximum temp. of external surface of beryllium reflector, °C	980
Average temp. of bases of radiating fins, °C	550
Maximum temp. of uranium dicarbide fuel elements, °C	1900
Total neutron flux in core center, $n/(cm^2)(sec)$	10^{13}
Total neutron flux at reactor-core boundary, $n/(cm^2)(sec)$	7×10^{12}
Neutron leakage from reactor, $n/(cm^2)(sec)$	3×10^{11}

One of the important aspects of a number of theoretical and experimental investigations done was the study of the operability of the thermoelectric elements in neutron and gamma fluxes. Repeated prolonged tests of thermoelectric elements in the loops of the reactor at integral neutron fluxes of 3×10^{19} thermal n/cm^2 revealed that the variation in the basic properties of the thermoelectric elements was within permissible limits.

In test-stand measurements thermal-energy characteristics were measured continuously with 53 tungsten-rhenium and 86 chromel-alumel thermocouples inserted in the reactor and the conversion device. The electrical output was determined at maximum power, the installation being tested for a total of approximately 100 hr during the test. The thermoelectromotive force of the device remained constant.

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Section

VII

Isotopes and Radiation Technology

Applications in Medicine, Biology, and Agriculture

Advances in the Use of Isotopes and Radiation Sources in Medicine

By E. H. Belcher,* M. Cohen,* R. A. Dudley,* H. G. Parker,† K. C. Tsien,* and H. Vetter*

Paper 880 (abridged)

Every year some 4000 scientific publications appear on medical uses of isotopes, including those on fundamental medical research, with about 1500 on diagnostic and therapeutic applications alone. Many survey papers and reports on new findings were given at the Second Geneva Conference, and this paper is confined to a review, albeit superficial, of developments since then.‡

There are only a few countries — certainly not more than 20 — where isotopes have not yet been introduced. The growth indicated by the increase in the number of licenses granted for medical use (Fig. 880-1) and in shipments (Fig. 880-2) of isotopes in some countries is thought to be indicative of the world situation. The great increase in availability of radioisotopes and labeled compounds for medical and other uses is indicated by the increase in entries in succeeding issues of the International Atomic Energy Agency (IAEA) International Directory of Isotopes:

Edition	Suppliers	No. of isotopes and labeled compounds (excluding stable)	
		Items	Entries
1st (1959)	44	1400	5,600
2nd (1962)	64	1950	7,500
3rd (1964)	83	2950	12,300

A number of countries have acquired research reactors and now produce the medically important isotopes ^{131}I, ^{32}P, and ^{198}Au for both their own use and for export. These reactors make the use of short-lived isotopes, e.g., ^{24}Na and ^{42}K, economic.

Nuclear medicine is beginning to be recognized as a specialty, and the number of those who still consider isotopes as a tool, like a microscope, that can be used by any medical doctor is rapidly diminishing. Chairs of nuclear medicine have been established in many medical faculties. National and multinational societies of nuclear medicine have been organized in Europe, the United States, Asia, and Latin America, and their several thousand

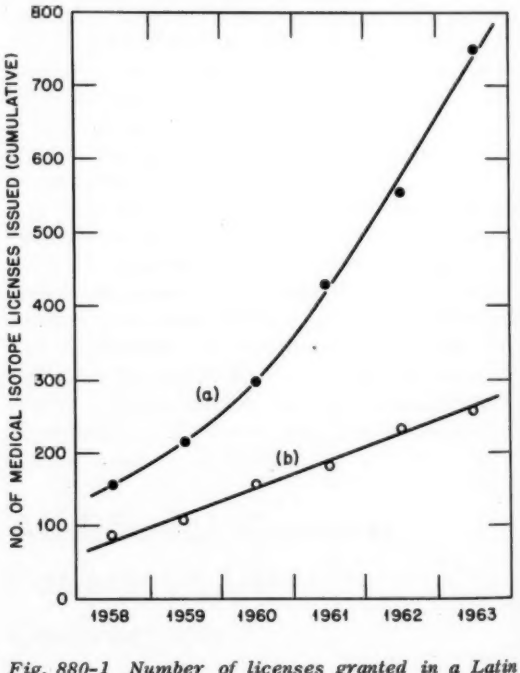

Fig. 880-1 Number of licenses granted in a Latin American country to physicians (a) for use of radioisotopes and (b) for therapy only.

*International Atomic Energy Agency, Vienna.

†World Health Organization, Geneva.

‡The applications of isotopes in medicine were reviewed in *Isotopes Radiation Technol.*, 1(1): 97-113 (Fall 1963).

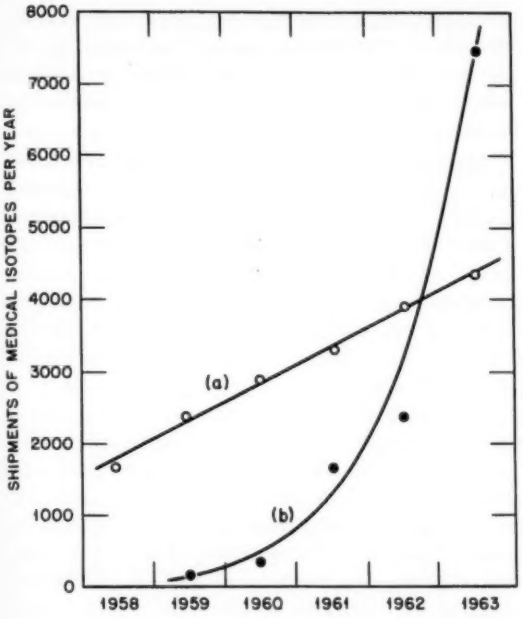

Fig. 880-2 Number of annual shipments of radioisotopes and labeled compounds for medical use by (a) a European and (b) an Asian producer.

members publish their own specialized journals. There can be little doubt that nuclear medicine will take the same course as radiology in becoming a medical discipline in its own right.

Diagnosis

ISOTOPES AND LABELED COMPOUNDS

The diagnostic use of some radioisotopes whose short physical half-lives make transport over long distances uneconomical is now feasible. The "milking"* method long used to obtain short-lived radon from long-lived radium is being used to produce, for example, 2.3-hr ^{132}I. This isotope, which is useful in thyroid-function tests, blood-volume measurements, and circulation studies, is a decay product of 78-hr ^{132}Te. The so-called "cow" of ^{132}Te, which is shipped to the consumer, continues as a source of ^{132}I for several weeks. The short-lived ^{26}Al, ^{137}Ba, ^{68}Ga, ^{140}La, and ^{99m}Tc can be obtained similarly.

With small cyclotrons being established at more universities and medical research institutions, we can expect an increasing use of cyclotron-produced neutron-deficient radioisotopes in medical work. An example is 2.1-min ^{15}O, whose use in cardiac- and pulmonary-function studies is a technical and organizational tour de force.

Great interest has recently been shown in the use of isotopes that emit low-energy gamma rays, e.g., ^{125}I, ^{147}Pm, ^{99m}Tc, and ^{197}Hg. This lower energy permits lighter and more efficient collimated scintillation counters to be used in uptake and localization studies in vivo. In many applications ^{125}I is replacing ^{131}I, not so much because of its low photon energy, although it has advantage in certain scintigraphic studies, but because the absence of particulate radiation lowers the radiation dose to the patient and the longer half-life permits longer storage of labeled compounds. Some promise is held for ^{125}I and ^{147}Pm as radiation sources in portable radiographic equipment.

Much progress has also been made in producing new labeled compounds and labeled compounds of higher specific activity. This is particularly true of tritium-labeled compounds, where the Wilzbach technique often gives specific activities of curies per millimole. The latest edition of the IAEA Directory lists some 600 commercially available tritium-labeled compounds.

EQUIPMENT AND TECHNIQUES

The most significant general advance in this field has been the introduction of fully transistorized electronic equipment into everyday medical isotope work. The availability of such equipment is particularly important in the developing countries because it is likely to stand up to rough conditions better than equipment with thermionic tubes and because printing circuits incorporating transistors can be readily replaced in the event of breakdown. Equipment manufacturers are trying to set up efficient maintenance and repair services throughout the world so that scalers, pulse-height analyzers, rate meters, etc., can easily be kept in good working order.

For in vitro measurement of low-energy beta emitters, such as tritium, ^{14}C, ^{35}S, and ^{45}Ca, liquid scintillation counting has become routine.

*This subject was reviewed by M. W. Greene, R. F. Doering, and M. Hillman, Milking Systems: Status of the Art, *Isotopes Radiation Technol.*, 1(2): 152-4, 158-9 (Winter 1963–1964).

Available equipment is highly automated, counting efficiency has been greatly increased, and self-quenching of colored samples, such as urine and plasma, has largely ceased to be a problem. In these counters and in solid well type scintillation counters for gamma emitters, automatic sample changers with data printout have become available. Thus large numbers of samples can be counted overnight and technicians freed for other work.

Considerable effort has been made to improve the sensitivity and versatility of scanning equipment for automatically recording the distribution of gamma emitters in the body. Commercial conventional scanning machines with moving detectors contain a scintillation counter with a 5-in. sodium iodide crystal and large multihole focusing collimators, which greatly improve depth resolution. Information can be stored on magnetic tape, and photorecording or closed-circuit-television techniques can be used to emphasize potentially significant information by discarding unwanted information. The recording of isotope distribution with stationary detectors has also made considerable progress with the development of so-called "scintillation cameras." Their advantage over conventional scanning machines is their ability to record changes within minutes.

Whole-body counters developed for radiation-protection work can also be used for measuring absorption and retention of small doses of radioisotopes in diagnostic studies. Their use allows a decrease in the radiation dose to the patient and eliminates the need for collection of human excreta. According to a recent IAEA survey, more than 60% of the whole-body counters now in operation are also used for clinical research. Many counting systems developed for radiation-protection work are too sensitive for diagnostic applications, and counters are being developed which are simpler and cheaper but still adequate for clinical work.

Many advances in in vivo measurement techniques are due to progress in the production of large scintillation detectors. Sodium iodide crystals up to 11 in. in diameter and 5 in. thick are now available. Cesium iodide crystals, offering a somewhat greater sensitivity per unit volume, have become available in smaller dimensions. Calcium iodide crystals have recently shown promise for detecting lower energy radiation with higher resolution but are still in the experimental stage. A most exciting recent development is solid-state radiation detectors. They are still too insensitive and too difficult to use for widespread application in medical work, but their excellent energy resolution holds great promise.

Electronic computers are being used more in medical isotope work. Analog computers analyze data in radioactive tracer studies and determine such parameters as pool sizes and rates of synthesis and degradation; digital computers appear to be particularly suited to analysis of complex gamma-ray spectrums recorded in whole-body counting or neutron-activation analysis.

Neutron-activation analysis is not strictly a medical application of isotopes and is in a very early stage of development. However, it will undoubtedly find many applications in medicine, particularly in determinating trace elements in human tissues or body fluids. Arsenic, iodine, and sodium can be determined rather easily; manganese and zinc require more elaborate procedures.

ADVANCES IN APPLICATIONS

Improvements in medical diagnostic uses of radioisotopes have proceeded along the expected lines: simplification of techniques, reduction of radiation dose, choice of the most reproducible tests, increased reliability of isotopically labeled materials and counting equipment, and study of a wider variety of organs and conditions by isotopic methods, particularly by gamma-ray scintillation scanning and scintillation-camera techniques.

The isotope used in greatest quantity is still ^{131}I, and the 24-hr thyroid uptake test with it is still the most common isotopic thyroid-function test. The use of ^{132}I, although a well-developed procedure that greatly decreases the radiation dose to the patient, has not displaced the use of ^{131}I in more than a few centers.* Iodine-125 has recently become available at competitive prices and will undoubtedly be used increasingly for thyroid tests.*

Widespread exchanges of standards and of information on methods of measurement in vivo have increased the accuracy of radioiodine-

*Iodine-132 was discussed briefly in *Isotopes Radiation Technol.*, 2(1): 82-5 (Fall 1964) and ^{125}I in 1(2): 143-6, 155-7 (Winter 1963–1964); 1(4): 343-5 (Summer 1964); 2(1): 67-70 (Fall 1964).

uptake measurements but have also disclosed rather wide variations in results reported by various clinical isotope laboratories (Fig. 880-3). Recent improvements in commercial equipment for measurements in vivo will make it easier to achieve more uniform results in the future.

The radioactive triiodothyronine test of thyroid function, which requires no administration of radioactivity to the patient, is now widely used, especially for screening. Substitution of a resin for the red cells originally used has further simplified it.

The radioisotope renogram, a kidney-function test that involves administration of a labeled substance selectively excreted in the urine and external recording of its passage through the kidneys by detectors placed over these organs, has become well established. Orthoiodohippurate labeled with ^{131}I, and more recently with ^{125}I, has become generally available; it is more selectively excreted by the kidney and has a lower uptake in the liver than the labeled Diodrast first available. Radioisotope renography is still semiempirical, as reflected by the wide variety of methods of data analysis in the literature. However, this situation is improving.

Renograms used with intravenous pyelograms (the standard contrast X-ray examination of the kidneys) have proved very helpful in selecting hypertensive patients for study by aortography and divided renal-function tests. This is an excellent example of a radioisotope test fulfilling its promise as a simple screening test since it often eliminates the necessity for more extensive procedures that carry much greater risk to the patient.

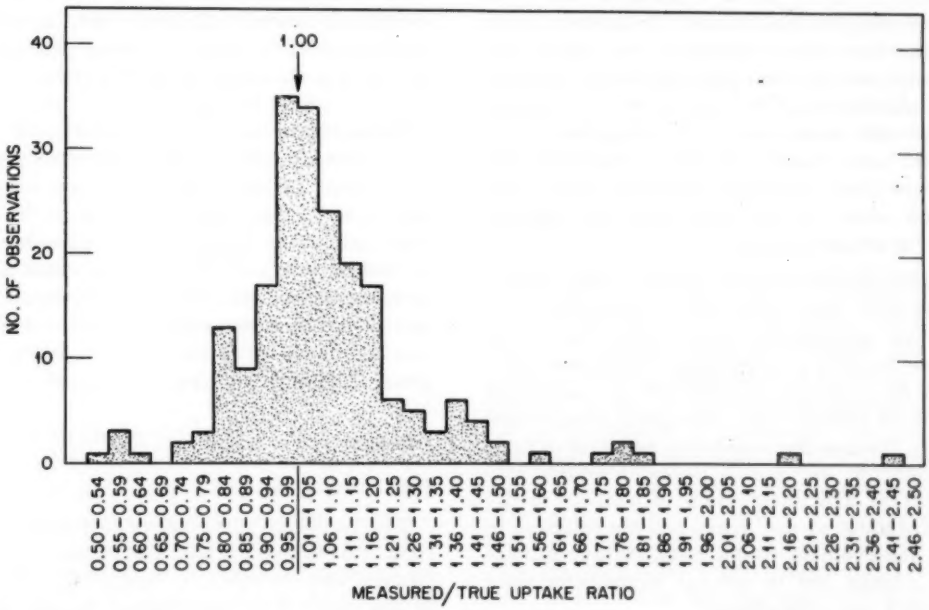

Fig. 880-3 Results of 219 calibrations of thyroid radioiodine-uptake measurements made in 36 medical isotope laboratories by an IAEA expert. A third of all observed values were more than 10% in error.

Gamma-ray scintillation scanning for determining the distribution of radioactive material in the body has made considerable progress. Scintillation scanning of thyroid, liver, and brain has spread, with the development of good commercial equipment, into many large general hospitals. In liver scanning valuable information can be obtained from a number of scans made in different planes. Gamma-camera techniques, in which the time required for a single scan is much reduced, permit such studies to be done routinely.

Theoretically, the best resolution for brain-tumor detection is probably provided by positron-scanning and positron-camera techniques. Although these methods have been well established in a few centers, they are not yet widely used because of the complexity of the equipment and the high cost of the positron-emitting isotopes. Brain scanning is presently being done with other equally useful gamma-emitting isotopes and a variety of recording techniques. The most widely used compounds are still ^{203}Hg-labeled Neohydrin and ^{131}I-labeled serum albumin. Since brain scanning can provide information not otherwise obtainable and since it is free from risk, it is being used as a screening test for more extensive diagnostic procedures even where clinical indications are relatively few. Scanning involves even less radiation dose to the brain than the common radiographic procedures.

Techniques for scanning kidney, bone, spleen, and pancreas have also proved clinically valuable. The gamma-ray scintillation scans promise to give a continuous visual presentation of the passage of radioisotopic materials through the kidneys with reasonably small doses and may replace the renogram. Scanning of lung, bone marrow, and lymph nodes is moving into the general armamentarium of nuclear medicine. Scanning of the heart and the stomach is a new achievement in the visualization of individual organs but is not yet competitive with standard X-ray procedures.

In clinical hematology ^{59}Fe and ^{51}Cr are being used increasingly for studies of red-cell production and life span, blood loss, gastrointestinal iron absorption, etc. For measuring vitamin B_{12} absorption in pernicious anemia and related conditions, ^{57}Co- or ^{58}Co-labeled vitamin B_{12} is now commonly used in place of the ^{60}Co-labeled vitamin. Cobalt-57 is especially useful because of its long shelf-life, good gamma-counting efficiency, and very low radiation dose to the patient due to the absence of particulate radiation. The newer technology has allowed quantity production of this particular isotope for medical uses.

A variety of other procedures have proved themselves in clinical work, e.g., the use of ^{24}Na- or ^{132}I-labeled albumin for placenta localization, the use of ^{131}I-labeled triolein and oleic acid for gastrointestinal fat-absorption tests, and the measurement of cardiac output by isotopic techniques. The radioactive gases ^{85}Kr and ^{133}Xe are evoking interest among clinicians for studies of muscle and cerebral blood flow and lung functions and are emerging from clinical research laboratories into regular use in larger medical centers.

Tritium and ^{14}C incorporated into an ever-increasing number of organic molecules are useful in biological and medical research, but clinical diagnostic uses are still few. Concern about administration to humans of compounds containing these long-lived radioactive atoms appears to have diminished somewhat, and many new research applications for them may soon be ready for wider use.

Methods for analysis of breath and excreta after administration of ^{14}C-labeled molecules have been further improved, and experience has been gained with a few more ^{14}C-labeled materials. The application of these techniques to studies with various labeled compounds, such as amino acids, will permit more detailed study of intermediary metabolism. In pernicious anemia and sprue, such studies have already shown their usefulness as simple diagnostic tests.

Therapy

SYSTEMIC ADMINISTRATION

The majority of therapeutic procedures relying on systemic administration of a radioisotope use its selective metabolic localization in the organ to be irradiated. No new developments in this area have occurred since 1958. Oral or intravenous ^{32}P therapy is still the method of choice for treatment of polycythemia vera. Phosphorus-32 is also frequently used in treating chronic leukemias. However, some hematologists prefer to use colloidal ^{198}Au, mainly because it is not excreted and thus problems of radioactive waste disposal do not arise. Iodine-131 continues to be widely used in therapy of thyroid cancer and thyrotoxicosis. However, in the last two years, careful analysis of long-term observations on patients treated for thyrotoxicosis has revealed the disquieting fact that permanent hypofunction of the thyroid gland may develop many years after ^{131}I therapy. In some centers as many as 50% of the patients appear to be so affected 10 years after successful therapy of thyrotoxicosis. Although other centers report figures very much less dramatic and, in any case, the condition can be controlled relatively easily by replacement therapy,

the whole situation requires thorough reassessment.

Many new methods of nonmetabolic localization of radioactive material in tissue to be irradiated have been attempted with varying success. If tissue is injured (e.g., by invasive growth of a malignant tumor), the coagulation system that converts fibrinogen to fibrin will be activated and protein will be deposited. Intravenous injection of ^{131}I-labeled fibrinogen or antibody to fibrin results in selective concentration of radioactivity in tumor tissue to an extent that suggests that this method might be used successfully in treating certain types of human cancer.

Lipiodol is an iodine-containing radiological contrast medium that can be labeled with ^{131}I to a high specific activity. If given intralymphatically it is removed by the lymph nodes, which can thereby be substantially irradiated. This form of therapy promises to be effective in treating metastatic involvement of lymph nodes in patients with cancer, particularly in the pelvic region.

If particles of a certain size are injected into the blood stream, their removal is a simple mechanical process. Particulate matter with a diameter of the order of 50 μ will not pass the small capillaries when injected intra-arterially, and doses as high as 100,000 rads may be delivered to a malignant tumor, particularly if it is highly vascularized. Ceramic microspheres labeled with ^{90}Y or resin particles labeled with ^{32}P are used.

INTERSTITIAL, INTRACAVITARY, AND SURFACE THERAPY

Considerable progress has been made in the use of artificial radioisotopes in place of radium and radon seeds for interstitial, intracavitary, and surface therapy. In many procedures radium can be replaced by ^{137}Cs, and an IAEA panel of experts recently recommended that newly established institutes or institutes with inadequate or unsuitable radium stocks use ^{137}Cs sources instead of radium. Cesium-137 rods for intracavitary application can be purchased in five different strengths (from 13 to 65 μc), and ^{137}Cs needles for interstitial therapy are available in standardized dimensions and activity, all at an initial cost much less than that of comparable radium sources. There is no possibility of gas leakage and, even if leakage of solid material occurred, the isotope is much less toxic than radium. On the other hand, its half-life is relatively short, and sources must be replaced about every 10 years.

In interstitial therapy, where the dimensions and shape of the source must be adjusted to the anatomical characteristics of the tumor to be irradiated, radioactive wires of ^{60}Co, ^{182}Ta, or ^{192}Ir are preferable. Iridium-192 can be drawn to tough wire with a high degree of flexibility, and its lower gamma energy decreases problems of protection.

Decisive progress in application techniques has been made by the introduction of so-called "after-loading" methods. The placing of radioactive sources is divided into two stages: (1) insertion of unloaded tubes and applicators and (2) subsequent loading with radioactive sources. Thus time, unlimited by safety considerations, is available for inserting nylon or plastic tubes or hollow stainless-steel needles into the tumor in an optimal pattern with maximum accuracy. At any time thereafter and, if necessary, even at another hospital, radioactive sources of exactly the right number, strength, and shape can be loaded or unloaded in a few minutes, and thus the radiation exposure of medical personnel is reduced to a minimum. This technique combined with the use of ^{137}Cs instead of radium would be particularly effective in some developing countries where the incidence of cancer of the cervix is approaching 30% of all cancers seen, and 5-year cure rates are likely to be greater than 50% for the early stages.

For permanent interstitial implants, ^{198}Au seeds are now preferred since their uniform size makes for easy implantation by mechanical devices. For irradiation of small volumes of tissue, such as the pituitary, pellets of the pure beta emitter ^{90}Y are advantageous because they can deliver a very high but localized dose. Cobalt-60 in the form of moulages is being advocated for surface therapy, and beta-emitting surface applicators of ^{32}P or ^{90}Sr have become established for treatment of very superficial lesions, such as those affecting the eye.

RADIOISOTOPE TELETHERAPY

The high energy of ^{60}Co gamma rays offers many advantages over the medium (180 to 250 kev) energy of the radiation produced by X-ray machines. Cobalt-60 gamma rays penetrate deeper into tissue and spare the skin, their differential absorption in bone is small, and deep-seated tumors can be treated more

effectively and with less discomfort to the patient. Cobalt-60 units are also more dependable. The rapid increase in the number of radiocobalt units in operation, which was apparent in 1958, has continued (Fig. 880-4), and many units have been installed in Europe, Japan, the USSR, and, more recently, in the developing countries.

The trend toward larger units has also continued. Units embodying 5000 curies of ^{60}Co are now in use. These units allow longer treatment distances as well as shorter irradiation times with a larger number of patients treated per day. Progress in producing ^{60}Co with higher specific activity has enabled this increase in source strength to be achieved without increasing the dimensions of the source, and new methods of producing ^{60}Co now under development promise to yield sources of still higher specific activity.

The number of ^{137}Cs teletherapy units has increased much more slowly. The specific activity of ^{137}Cs will have to be much higher than it is at present if such sources are to be kept small enough to avoid a large penumbra and if their dimensions are to be kept within the range of those of present ^{60}Co sources. Because of the difference in gamma energy, the two radiation sources are not really comparable. It might well be that ^{137}Cs units will ultimately replace conventional X-ray therapy machines for treatment of lesions that do not require "supervoltage" radiation.

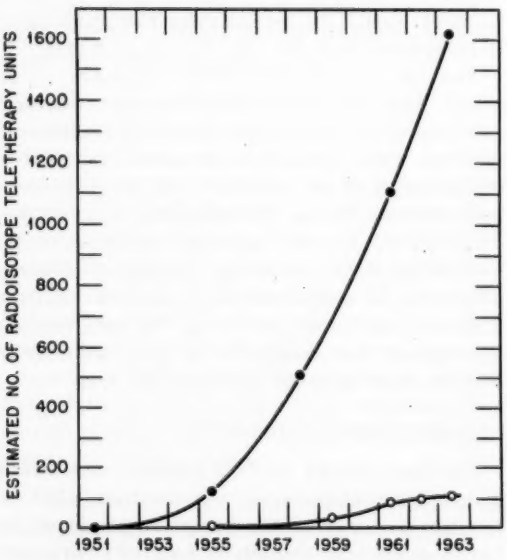

Fig. 880-4 Estimated number of radioisotope teletherapy units in operation throughout the world. ●, total. ○, ^{137}Cs.

The increased use of high-energy radiation for teletherapy has accentuated the need for more accurate determinations of the absorbed dose. Although the physical characteristics of ^{60}Co units are now well understood and such data as isodose curves and depth-dose tables are readily available, interest is shifting toward a better understanding of the effects of the individual anatomy of the patient on dose distribution. The patient is no longer considered as a rectangular tank of water, but allowance is beginning to be made for body shape and composition. Transverse topography is used increasingly to determine the exact topography of the region of interest in its relation to surrounding tissue, and transit doses and doses within the body cavities are measured to account for differences in absorption of radiation when it passes through air spaces or such tissues as lung. Beam-modifying devices are used increasingly to achieve the desired dose distribution. This type of work requires a great deal of mathematical calculation, particularly for complex radiation-field arrangements, and it is not surprising that digital computers are playing a more important role in accurate treatment planning.

REACTORS

At the Second Geneva Conference, considerable time was devoted to a discussion of the use of reactors for neutron-capture therapy. In theory this type of therapy is particularly promising in the treatment of brain tumors. Of the nuclides that produce alpha radiation following neutron capture, ^{10}B has the highest cross-section. A large number of boron-containing compounds have been tested for their selective concentration in brain tumors, and, following administration of such compounds, several series of patients were subjected to thermal-neutron radiation at the Brookhaven National Laboratory medical research reactor and the Massachusetts Institute of Technology research reactor. The clinical results obtained so far are rather disappointing, and many years of intensive research will be required before the use of reactors for radiotherapy will become practical.

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Separation and Purification of Certain Radioisotopes for Medical Use

By A. A. Abdel-Rassoul, H. Fouad, A. Abdel-Aziz, S. S. Wahba, and S. Mohammed*

Paper 457 (extracted)

New procedures were investigated for separating and purifying ^{35}S, ^{45}Ca, ^{46}Sc, ^{48}Sc, ^{111}Ag, and ^{199}Au and for preparing carbon suspensions containing ^{198}Au and ^{111}Ag.

Sulfur-35 (87 days), $^{35}Cl(n,p)^{35}S$

A method was developed for separating ^{35}S, produced by neutron irradiation of potassium chloride, from the ^{32}P produced simultaneously.

*Atomic Energy Establishment, United Arab Republic.

The sulfur and phosphorus are converted to sulfate and phosphate, respectively, and the sulfate is selectively sorbed on the basic anion-exchange resin Dowex 1 and quantitatively eluted with 0.1*M* HCl. The product has a radiochemical purity greater than 99.9%.

Calcium-45 (165 days), $^{48}Ti(n,\alpha)^{45}Ca$, and Scandium-46 (84 days), $^{46}Ti(n,p)^{46}Sc$

Carrier-free ^{45}Ca and ^{46}Sc are recovered from a solution of reactor-irradiated potassium fluorotitanate, K_2TiF_6, by sorption on the acidic anion-exchange resin Dowex 50. The ^{45}Ca is eluted first with 1*M* HCl, and then the ^{46}Sc with 4*M* HCl. The products are practically free from chemical residues and have radiochemical purities of greater than 99.9% and greater than 99%, respectively.

Scandium-48 (1.8 days), $^{51}V(n,\alpha)^{48}Sc$

From a hydrochloric acid solution of an irradiated V_2O_5 target, vanadium is sorbed on Dowex 1. The ^{48}Sc, complexed with oxalic acid, is recovered from the effluent with a radiochemical purity greater than 99%.

Gold-199 (3.14 days), $^{198}Pt(n,\gamma)^{199}Pt \xrightarrow{-\beta} {}^{199}Au$

Carrier-free ^{199}Au, formed from ^{198}Pt through the intermediate ^{199}Pt, was readily separated from the target solution by cocrystallization with NaCl. The process is still preliminary.

Silver-111 (7.5 days), $^{110}Pd(n,\gamma)^{111}Pd \xrightarrow{-\beta} {}^{111}Ag$

Irradiated palladium is dissolved, and the silver is carried from solution on sodium chloride. A saline solution of the ^{111}Ag may be used medically, or the silver may be electrodeposited on platinum and subsequently dissolved in nitric acid.

Gold-198 (2.7 days), $^{197}Au(n,\gamma)^{198}Au$, and Silver-111 on Carbon

A solution of irradiated gold, as $Na^{198}AuCl_4$, is adsorbed on carbon and reduced with ascorbic acid. Gold-198 can be similarly deposited at the same time to increase the time of effective action of such preparations. The particles are filtered off and suspended in a medium suitable for medical use, prepared from carboxymethyl cellulose, polyethylene glycol oleate, and sodium chloride.

Biological Disposition of Antibilharzial Antimony Drugs

Estimation of Antimony in Body Fluids and Parasites by Activation Analysis

By A. A. Abdel-Rassoul,* M. F. Abdel-Wahab,* A. R. Schulert,† H. G. Browne,† and M. M. Mansour†

Paper 841 (abstracted)

A neutron-activation procedure was used to determine antimony in body fluids and parasites after administration of antimony-containing drugs to patients with bilharziasis. Red blood cells, urine, and excreted *Schistosoma hema-*

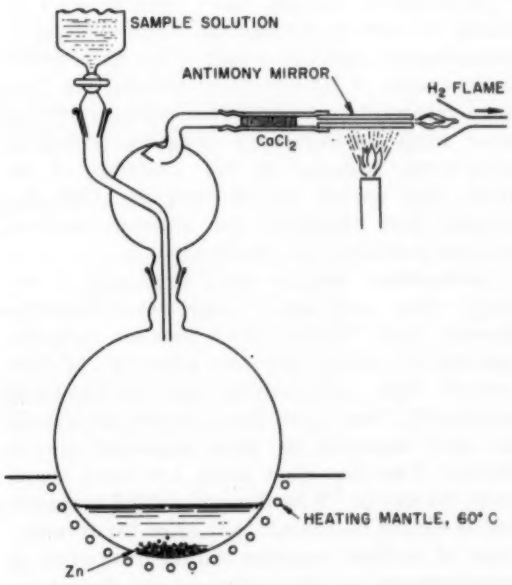

Fig. 841-1 Apparatus for separating antimony from irradiated biological samples.

*Atomic Energy Establishment, Inshas, United Arab Republic.

†U. S. Naval Medical Research Unit No. 3, Cairo, United Arab Republic.

tobium ova from patients and *S. mansoni* ova and male and female worms from infected hamsters were assayed.

The method can determine 0.005 μg of antimony with 10% or better accuracy. Ashed samples are irradiated 48 hr at a neutron flux of 1.3×10^{13} n/(cm^2)(sec). A weighed amount of the irradiated material, which contains ^{122}Sb and ^{124}Sb, and a suitable weight of Sb_2O_3 carrier are dissolved in concentrated sulfuric acid in the presence of potassium pyrosulfate. The solution is cooled, diluted to 30% H_2SO_4, and dropped onto zinc (Fig. 841-1). The SbH_3 in the off-gas is converted to antimony metal, which is deposited, dissolved in sulfuric acid, and counted.

Details of history and procedure and results on patients are given in the original paper.

Tracers in Biology

By Carlos Chagas*

Paper 874 (abridged)†

Radioactive isotopes have been used in all fields of modern biology but most usefully in biochemistry and physiology. The main metabolic cycles of intermediary metabolism have been untangled, and rates of numerous processes have been measured in complex biological structures. Because of the vastness of the field, this review is not complete. Only examples that illustrate the diverse uses of tracers in biology are presented.

Radioactive isotopes were first used in biology when only natural ones were available. Hevesy[1] used ^{210}Pb to solve a problem in plants. Impetus to tracer use was given by the discovery[2] that radioactivity can be produced artificially, thus providing a means for making the light elements, the most important ones in biology. Two important dates are those which mark the use of ^{32}P by Hevesy[3] and the preparation of tagged thyroxin.[4] Only after the development of nuclear reactors did the production of radioisotopes become sufficient for their general use and for progress in preparation of tagged molecules.

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†Original in French (translated into English as ORNL-tr-426).

Sulfolipid in Green Plants

By the use of radioactive tracers, an unexpected natural compound was detected and its structure determined without isolating it in chemically pure form. Benson et al.[5] cultivated photosynthetic organisms in a medium containing $^{35}SO_4^{2-}$, extracted the lipids, and separated them by paper chromatography followed with autoradiography. The radioactivity was concentrated in a band that should correspond to an unknown sulfolipid. The photosynthetic organism was then cultivated in a $^{14}CO_2$ atmosphere, and the lipid extract was subjected to chromatography, which gave the sulfolipid uniformly tagged with ^{14}C.

Acid hydrolysis of the sulfolipid gave glycerol, fatty acids, and an unknown compound that migrated very little on the paper and was twice as radioactive as the glycerol and therefore contains 2×3 carbon atoms = 6 carbon atoms. When the ^{35}S-tagged sulfolipid was hydrolyzed, all the radioactivity was found in this unknown compound, whose acidity corresponded to that of a sulfonate. It gave an osazone and a hydrazone and could be acetylated; it was shown to be glucose-6-sulfonate.

The radioactivity of fatty acids tagged with ^{14}C indicated the presence of 34 atoms of carbon. A plant sulfolipase gave a single fatty acid,[6] which was identified as linoleic acid (18 atoms of carbon). A second fatty acid liberated by acid hydrolysis was identified as palmitic acid (16 atoms of carbon). These facts, together with enzyme-activity evidence, established the formula shown in Fig. 874-1.

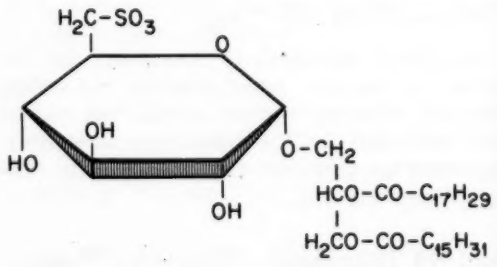

Fig. 874-1 Formula of the sulfolipid.

Activation Analysis

Activation analysis for determining trace elements made radioactive by neutron capture[7,8]

has become a complementary technique to paper chromatography.

Keynes and Lewis[9] measured the sodium and potassium contents of nerves at rest and of stimulated nerves and determined that stimulation is accompanied by an increase in the sodium content and a decrease in potassium. Comar et al.[10] determined the total thyroid iodine.

Activation analysis cannot be used with organic molecules because the activation usually liberates the element from its chemical bonds. However, in activation chromatography[11] the nonradioactive compounds are separated by bidimensional paper chromatography, and the chromatograph is then neutron irradiated. The activatable impurities in the paper have short half-lives, and the decay is followed by measuring the radioactivity induced in the compounds that are to be determined and whose identities have been established by autoradiography. Phosphorylated compounds lend themselves very well to this kind of analysis. Many compounds that are not directly activatable can be determined after conversion to derivatives containing an activatable element, such as bromine. Thus organic acids can be determined in the form of *p*-bromophenacylated esters, the monosaccharides and cetonic acid as *p*-bromophenyl hydrazones, and amino acids as *p*-bromobenzenesulfonyl amides.[12]

Assimilation of Carbon in Photosynthesis and Chemoautotrophy

Chlorophyllic assimilation comprises two distinct processes: (1) conversion of solar energy to chemical energy in the chloroplasts and (2) formation of glucides and various organic compounds from atmospheric CO_2. Study of the first process and its expression in physicochemical and biochemical terms is difficult and is in its initial stages. Study of the second process may be considered successful along broad lines and has developed significant information on the photosynthesis cycle.[13]

Bidimensional paper chromatography can be combined with autoradiography to show the presence of radioactive compounds formed from a tagged substrate.[14] Thus the biochemical reactions of an intact organism are studied kinetically, and the effects of sudden changes in external conditions on the tagging of the metabolites are shown. The complexity of the intermediate steps of the photosynthesis cycle (Fig. 874-2) is such that it would have been practically impossible to study them without radioactive tracers. In each cycle a molecule of CO_2 is fixed, and a molecule of ribulose diphosphate is resynthesized; six cycles are required to form one hexose and 12 to form a disaccharide. When a green plant is exposed to tagged CO_2, the first stable product formed is 3-phosphoglyceric acid.[15] Chemical degradation

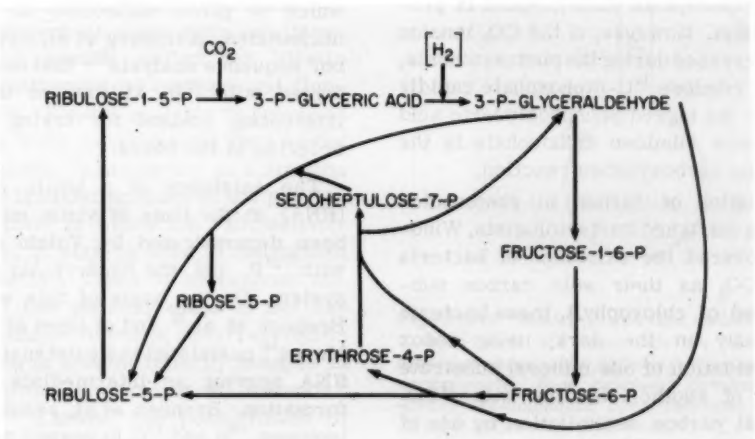

Fig. 874-2 The CO_2 assimilation cycle.

of this compound has shown that the radioactivity is incorporated first in the carboxyl group and then in the alpha and beta carbon atoms. The alpha and beta carbon atoms do not come directly from $^{14}CO_2$ but from ribulose diphosphate, whose presence in the green plant has been established.[16] The CO_2 is fixed by carboxylation of ribulose diphosphate, which is eventually broken into two phosphoglyceric acid molecules, only one of which contains the newly incorporated ^{14}C. The 3-phosphoglyceric acid is reduced to 3-phosphoglyceric aldehyde and dihydroxyacetone phosphate, which is condensed to fructose diphosphate.[17] Actually the tagging of hexoses is more complicated[18] since the C_4 is selectively tagged over the C_3 and then the C_1 and C_2 over the C_5 and C_6. Five triosephosphates form three pentosephosphates.[13]

A cyclic relation between the phosphoglyceric acid and the ribulose diphosphate during chlorophyllic assimilation has been established: If the light is extinguished, the concentration of ribulose diphosphate rapidly decreases[19] because the carboxylation continues, but phosphorylation of ribulose-5-phosphate ceases for lack of adenosine triphosphate (ATP), which is produced in the light. However, if the CO_2 tension is suddenly decreased during the photosynthesis, tagging of the ribulose ^{14}C-diphosphate rapidly increases, and the tagged phosphoglyceric acid decreases[20] since ribulose diphosphate is the substrate for the carboxylation reaction.

The assimilation of carbon in chemoautotrophy has long intrigued bacteriologists. Winogradski[21] discovered the existence of bacteria that require CO_2 as their sole carbon substrate. Deprived of chlorophyll, these bacteria develop perfectly in the dark, using redox energy from oxidation of one mineral substrate and reduction of another. Studies with $^{14}CO_2$ have shown that carbon assimilation by one of these bacteria, *Thiobacillus thiooxidans*, is identical to that of chlorophyllic assimilation.[22,23] It has been demonstrated that ATP is the first energy-rich product formed. It has not been possible, however, to establish the stages preceding ATP formation.[24]

The Genetic Code and Protein Synthesis

Many investigations on control of protein synthesis by the genetic code would have been impossible without stable and radioactive isotopes. Watson and Crick[25] are credited with the concept of the genetic code, which is one of the most stimulating ideas in modern biology. The role of deoxyribonucleic acid (DNA) in the transmission of genetic information was shown in the work of Avery et al.[26] on the pneumococcus transformation principle. Irrefutable proof was brought by Hershey and Chase,[27] who tagged phage T_2 with ^{32}P for DNA and ^{35}S for proteins and then infected bacteria. They found the ^{32}P in the newly synthesized phage to the exclusion of ^{35}S and thus established the importance of DNA and eliminated protein participation in the transmission of genetic information. Meselson and Stahl[28] used stable nitrogen to study the mechanism of DNA replication; bacteria were cultivated in a medium containing $^{15}NH_4Cl$ as the only nitrogen food and then transferred to a medium containing ordinary ammonium chloride. By centrifugation in a cesium density gradient, the DNA tagged with ^{15}N was separated from ordinary DNA.

Ochoa and Heppel[29] prepared synthetic polyribonucleotides, and Kornberg et al.[30] obtained synthetic polydeoxyribonucleotides by an enzymatic method. To determine the frequency with which a given nucleotide is bound to other nucleotides, Kornberg et al. developed a neighbor sequence analysis[30a] that uses the nucleotide tagged with ^{32}P. At present this is the most interesting method for trying to untangle the ordering of the bases.

The existence of a labile ribonucleic acid (RNA) at the time of virus multiplication has been demonstrated by Volkin and Astrachan[31] with ^{32}P and the *Escherichia coli* – phage T_2 system. On the basis of this work and that of Brenner et al.,[32] and of Gros et al.,[33] Jacob and Monod[34] postulated the existence of a messenger RNA serving as intermediate for genetic information. Brenner et al. associated the stable isotopes ^{15}N and ^{13}C to permit separation of the nucleic acids by a density gradient to the radioactive isotopes ^{14}C and ^{32}P used to tag the nucleic acids and ^{35}S used to tag the proteins. They demonstrated that after an infection of *E. coli* by the phage T_4, a labile RNA is formed which attaches to the RNA of the ribosomes present before the infection. Gros et al. extracted this labile RNA and determined its sedimentation constant.

The polyribonucleotides prepared by Ochoa et al.[29] can serve as a model for understanding the role of the RNA messenger. A synthetic

polyuridylic messenger induces the formation of a polyphenyl alanine chain in a system containing bacterial ribosomes (Nirenberg and Matthaei[34a] and Ochoa et al.[35]). Jukes[35a] tried to compare the mutations carried on a single amino acid to the changes of a single base in certain RNA messenger triplets. Weiss and Nakamoto[36] polymerized ribonucleotides in the presence of DNA, and RNA was formed by the DNA primer.

The activity of DNA and RNA can be shown in the polytenic chromosomes of the salivary glands of Chironomides and *Rhynchosciara.* Using ^{3}H-tagged thymine, cytidine, and uridine, Ficq and Pavan[37] showed by autoradiography that DNA and RNA are synthesized in chromosome puffs and that the activity varies from one point of the chromosome to another with the age of the larvae.

Kinetic Analysis

The first studies with radioactive tracers were made in an attempt to solve simple problems, and the most important idea developed was that of renewal and dynamic equilibrium.[38] As the problems became more complex, interpretation of the results called for kinetic analysis, i.e., mathematical analysis and elaboration of models. Kinetic analysis is proposed for interpreting such experimental data as the variation of specific radioactivity as a function of time and for translating them to the physiological parameters in which the radioactivity does not appear. It may be used to distinguish between two distinct types of movement: exchange between two phases, which is not accompanied by changes in mass, and irreversible movement, which is accompanied by changes in mass (e.g., the process of excretion). Solomon[39] and Robertson[40] showed the possibilities of kinetic analysis with several theoretical examples.

Since change in the specific radioactivity as a function of time can be expressed as the total of the exponential functions, the existence of compartments in the models is suggested. The system must comprise a number of compartments equal to the number of exponential functions to the point of becoming unsolvable. If there are more than two compartments, several models can be visualized which differ in compartment arrangement and connection by means of the exchange process and by the transfer in and out of the compartment(s). With only two compartments, if the exchange phenomenons are unequivocal, there are still numerous combinations of the disposition of one or several influxes and irreversible effluxes. All these models are equally acceptable from the mathematical point of view, and we choose that which agrees best with known data.

In the rat the balance method combined with interpretation of the decrease in the specific radioactivity of serum calcium after intravenous injection of $^{45}CaCl_2$ led Milhaud and coworkers[41] and Aubert and Milhaud[42] to establish a model with two compartments (Fig. 874-3) and to measure the chief parameters of calcium metabolism. This model is used for studying

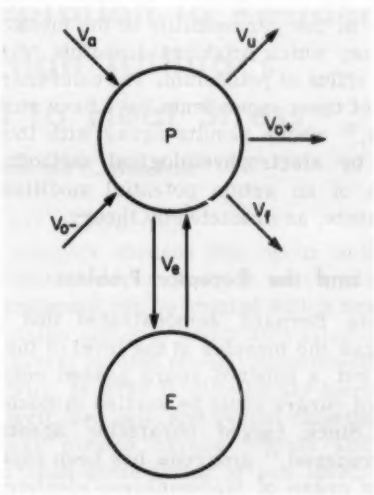

Fig. 874-3 Model of calcium metabolism in the rat. Compartment P: blood calcium, extracellular calcium, exchangeable soft-tissue calcium, and rapidly exchangeable bone calcium; compartment E: more slowly exchangeable bone calcium; V_e, rate of exchange between compartments; V_{o+}, bone anabolism; V_{o-}, bone catabolism; V_f, excretion of fecal endogenous calcium; and V_a, calcium absorbed in digestion. [V_u is excretion of calcium.]

the effect of treatment with vitamin D_2 (Ref. 41) and cortisone,[43] effect of hypophysectomy and treatment with the somatic hormone and thyroxin,[44] effect of thyroid and parathyroid glands[45] and age[46] on bone and calcium metabolism in experimental rickets, and relations between the various processes in the normal rat.[47]

In the case of iron metabolism, which is particularly important for the study of erythro-

kinetics, the initial model had a single compartment,[48] then two;[49] its actual simplified form has three.[50]

These models have rendered great service in elucidating the physiopathological mechanisms of interest in the metabolism of calcium and iron and in erythropoiesis. They are of particular interest for evaluating the importance of incorporating the radioelements in the biosphere.

Nerve Influx and Ionic Movements

Certain aspects of the ionic theory of Hodgkin and Huxley[51] by which they explain the formation and propagation of nerve action potential have been confirmed by the use of tagged sodium and potassium. This theory assumes a change in the permeability of the membrane of the axon, which produces an influx of sodium and an efflux of potassium. The chief characteristics of these movements have been studied by Keynes,[52] whose results agree with those obtained by electrophysiological methods. Production of an action potential modifies these movements, as predicted by theory.

Curare and the Receptor Problem

Claude Bernard demonstrated that curare paralyzes the muscles at the level of the motor plate, but a hundred years passed before the action of curare could be studied in biochemical terms. Since tagged curarizing agents have been prepared,[53] progress has been rapid. The electric organ of *Electrophorus electricus* behaves like a huge motor plate, curarization of which abolishes the potential.[54] Tagged curarizing agent was first injected in the living animal by Hasson and Chagas[55] and by Chagas.[56] When the electric response had been abolished and the electric organ extracted, the radioactivity was found bound to a fraction of increased molecular weight,[57] which has been identified as a mucopolysaccharide acid.[58] Since curare is not metabolized in the organism, the relation between curarizing agents and the purified mucopolysaccharide acid can be studied in vitro.[59]

If we use the smallest intact unit of electric tissue, the isolated electroplate,[60] we can follow the process of fixation of tagged curare and relate it to the progressive abolition of the transmission of the nerve influx.[61] We can relate the defixation of curare to the progressive recovery of the influx transmission.[61] Autoradiography has shown that curares are fixed in the electric organ in vivo in the synapses and in the connecting tissue.[62] Waser[63] obtained semiquantitative images that illustrate the inhibition of the cholinergic transmission in the rat diaphragm.

Thus the use of tagged curarizing agents permitted solving a fundamental pharmacological problem, that of the specific receptors of pharmacologic agents.[64]

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Radioisotope Studies of Adaptation to Residence at High Altitudes and to Heat Stress

By J. Metz and C. Wyndham*

Paper 853 (abstracted)

The complex changes that occur in the fluid compartments of the body in man's adaptation to environment can be studied with a new order of precision and over short time intervals with radioactively labeled normal constituents. Persons living at 5740 ft have higher hemoglobin, hematocrit, red cell, and serum chloride values but lower plasma carbon dioxide and serum sodium than those living at sea level. Blood-volume changes responsible for these differences were studied with red-cell-volume measurements made with ^{51}Cr, plasma-volume measurements made with radioiodinated human serum albumin, and total-body-water measurements made with tritiated water.

The reaction of the body to a single acute exposure to severe heat was examined. The absence of plasma-volume changes was shown with radioiodinated human serum albumin, and changes in total body water were studied with tritiated water.

Original paper gives background and details of results.

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Advances in the Use of Isotopes and Radiation Sources in Agriculture and Food Science

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Paper 876 (abridged)

Since the Second Geneva Conference, progress has been steady in the application of nuclear techniques in all fields. Radioactivation analysis appears to have considerable potential for determining trace elements in soils and biological materials although many problems still must be solved before its establishment on a routine basis. In work on food irradiation, the problems of wholesomeness, palatability, and economics are steadily being overcome. And the development of reactors for desalination of water for crop irrigation is of great interest although the economics have not been solved.

Tracers and Small Radiation Sources

SOILS

Chemistry. The isotope-dilution law has been applied to equilibrium soil solutions for determination of ion-exchange equilibriums. Not only can systems of low salt and moisture contents be studied, but they can also be analyzed without having to determine absorbed elements by the use of empirical extraction techniques, which introduce large errors.[1–3]

The introduction of carrier-free or high-specific-activity radioisotopes to equilibrium soil systems has permitted determination of the rates at which reserves can exchange with elements in the soil solution. The application of isotope techniques to ion-exchange kinetics has yielded important information on the nature of cation- and anion-exchange mechanisms in soil systems.[2,4–12]

The magnitude of the nutrient reserve in the soil, the so-called "labile pool," has been determined in both soil suspensions and soils of normal moisture content; a plant is used to sample the solution, and the specific activity is determined (Refs. 2, 9, and 13–19).

Fertility. Techniques using labeled fertilizers have been developed for comparing the availability of elements in different soils and fertilizers and for determining the best time for and methods of applying fertilizer, the residual value of fertilizers, and interactions between fertilizers. An exact meaning can now be given to the availability of elements in the soil, the availability of a particular fertilizer being used as a standard of comparison and the plant as the soil extractant (Refs. 2, 3, 9, 16, and 20–25).

Physics. Measurement of soil moisture by the neutron method is well established[26–28] although the initial cost of the equipment is a hindrance to wider use. Substantial advantages of this method include direct and rapid determination in the field, repeated recording at any desired depth without further soil disturbance, instantaneous equilibration with soil moisture, and insensitivity of the method to soil salt concentration. Problems have centered around instrument calibration, and many papers have dealt with the efficiency of the method.[29–35] Studies have been made on water–air space relations in soils,[36] water availability for crops,[37] control of moisture in greenhouse studies,[38] and measurement of infiltration characteristics of soils.[39]

Soil density can be measured by counting gamma rays emitted from a source and backscattered by soil atoms to the detector. One study showed that the procedure was as precise as conventional soil core methods and more than three times as fast.[40] Gamma-irradiation measurement of soil moisture[41,42] has been used in research on unsaturated water flow in soils,[43,44] and the internal water status of plants in relation to soil moisture supply has been determined by beta-ray gaging.[45]

By investigation of the thermodynamics and kinetics of soil moisture with a ^{60}Co-ethylenediaminetetraacetic acid complex, the changes in the relation between fluxes of moisture through the liquid and gaseous phases under varying conditions have been followed.[46] The hydrophysical characteristics of peat have been studied with ^{35}S and ^{131}I as nonsorbable indicators.[47]

*International Atomic Energy Agency, Vienna.
†Food and Agriculture Organization of the United Nations, Rome.

PLANTS

Plant Physiology. Future basic studies on absorption of nutrient ions by plants have extended our knowledge of the process for almost all the major elements.[48–54] Absorption and loss of nutrients by leaves have also been studied.[55–57]

Substantial work has been done on translocation of assimilates (products manufactured by the plant) in tobacco,[58] soybeans,[59–62] wheat,[63,64] and sugar cane.[65,66] Carbon-11 (20.4 min) has been used directly after preparation in the cyclotron.[61] After decay of the activity, the same plant can be used again under different conditions. An ingenious technique for measuring lateral movement of assimilates in plants consists in exposing a honeydew leaf to ^{14}C and measuring the activity of an aphis colony feeding on the opposite side of the stem.[67–69] Studies have also been made on movement of inorganic ions in plants.[70–74]

As representative of the use of isotopes, particularly ^{14}C, in studies on synthesis and metabolism, we may note investigations of glucose utilization,[75,76] incorporation of proline into proteins,[77a,b] malonic acid biosynthesis,[78] acetate utilization,[79] and incorporation of magnesium into chlorophyll.[80]

Plant Pathology. Tracers have been used to study metabolism and physiology of diseased or injured plants,[81,82] translocation of fungicides,[83] and the fate of fungicides in the plant.[84] Spray deposits have been evaluated, including the spraying pattern and losses due to rainfall, with plant-protection chemicals made radioactive by neutron irradiation.[85] Distribution patterns have been investigated by autoradiography of the leaves. In studies with ^{14}C-labeled fungicides[86,87] and dosages as low as 1 ppm, fungal spores removed large amounts of toxicant in a short time, but large amounts were required to inhibit 50% of the spores.

Weeds. Elegant autoradiographic techniques have been developed[88] for precise study of the mobilities of labeled herbicides[89] and factors affecting translocation.[90,91] The effect of herbicides on metabolism has been studied with ^{14}C-labeled compounds.[92–95] Work on 2,2-dichloropropionic acid (DCPA or Dalapon) illustrates the varied approach possible with isotopes, with studies on penetration and translocation,[96] distribution and metabolism,[97] general physiological action,[98] and absorption from soil.[99]

Plant Breeding. The discovery that the natural mutation rate could be multiplied by irradiation opened up a vast new potential for the plant breeder,[100] and radiation has proved to be an important microsurgical tool for transferring an individual characteristic from one species to another.[101–103] Even when a desirable characteristic can be found in a wild variety, irradiation of the seeds of an adapted species can induce the particular mutation more quickly and economically than hybridization with the wild variety and selection by conventional breeding methods.[104,105]

Gamma fields have been constructed in many countries, the classical gamma source, ^{60}Co, being in many cases replaced with the better-suited ^{137}Cs. Research reactors have been constructed, which give the advantage of a neutron source for irradiation. The discovery that some chemicals are efficient mutagenic agents[106–108] has resulted in increased research activity and improved understanding of the nature of induced mutations[109] and methods for their successful induction.[110] In some crop plants a number of induced mutations have been localized on the chromosomes.[111] New methods of screening for mutations have been formulated and tested.[110,112,113]

Artificial induction of mutations may not be merely an increase in the rate of naturally occurring mutations; actually, real differences may exist in the spectrums of natural and artificially induced mutations.[114,115] Alteration of the environment prior to, during, and after irradiation may modify and direct the induced genetic changes and decrease the somatic damage.[116,117] The mutability of the specific gene locus for resistance to *Helminthosporium victoriae* in oats can be fundamentally increased by changes in the water of the seed prior to gamma irradiation.[118] Recent developments in knowledge of mutation spectrums of different kinds of radiations and chemical mutagens promise that combinations of mutagens with modifications of the pretreatment and post-treatment conditions of the plant material may soon be used to induce at will specific desirable gene and chromosome changes.[100]

Nuclear volume and chromosome number, particularly, are good indexes for radiosensitivity.[119,120]

Much of the initial skepticism toward the usefulness of induced mutations in plant-breed-

ing programs has disappeared as a result of better understanding of the mutation process, improved techniques of screening for mutants, and important practical results with the use of radiation-induced mutants in plant breeding.[121,122] Examples of these practical results are another new barley variety developed in Sweden,[121,123] a new peanut variety,[124] and a crown rust–resistant oat variety.[104] A single X-ray treatment of *Hibiscus sabdariffa* produced progenies with a changed photoperiod response, which enabled it to grow 1000 miles north of its natural habitat.[125]

ANIMALS

Nutrition. Following classical work on mineral metabolism,[126–128] numerous studies have been made on the relation between the apparent and true digestibility of minerals and their distribution in the body. For example, the true digestibility of magnesium in hay was determined by ^{28}Mg studies with sheep.[129–131]

Radioisotopes have been used to locate the part of the intestine where various nutrients are absorbed and to show the interference of an excess of one mineral with the digestibility of others.[132] Besides confirming previous knowledge of iron distribution in the body, tracer studies with pigs have shown that the entire intestinal tract may take part in iron absorption.[133]

Metabolism. Studies with labeled materials have shown the role of the liver in phospholipid synthesis and of phospholipids in fat metabolism and in the transport of fat to the mammary tissue.[134,135] Tracers have been extensively used in studies on the exchange of minerals between body fluid compartments and tissues. For example, through the use of ^{28}Mg, the total exchangeable and nonexchangeable body magnesium has been established,[129] and the conservation of magnesium in soft tissue and its depletion in bone during magnesium starvation has been shown.[136]

Milk Secretion. Early experiments with isotopes showed that casein is synthesized in mammary tissue from free amino acids taken up from the plasma.[137] Recent studies with ^{14}C-glucose and proxionate [sic; propionate?] showed that the mammary tissue also synthesizes amino acids from nonprotein sources.[138] Other tracer studies on milk-fat synthesis have shown many important facts about the origin of short- and long-chain fatty acids and glycerol.[139–141]

Reproduction. The time interval between the formation of a male gamete and its appearance in semen has been studied by injecting soluble phosphates, which subsequently become incorporated in DNA-^{32}P of the sperm.[142–144] Autoradiographic techniques and ^{3}H-thymidine as DNA precursors have made possible a more differentiated interpretation of the age of sperm in the ejaculate.[145] Resin spheres impregnated with ^{198}Au have been used to determine the exact speed of egg transport in the Fallopian tube.[146]

Pathology. Since earlier methods of using radioiodine for studies on the thyroid gland[147–150] are not suitable for clinical investigations, a new and indirect way has been found in which a stabilized blood sample is mixed with labeled triiodothyronine (T_3). The rate of uptake of this compound by the erythrocyte reflects the functional state of the thyroid gland.[151,152] This rapid test has been used on a large scale for measuring thyroid states in livestock.[153]

The Rose Bengal-^{131}I liver-function test has been used in sheep to show the depression of rate of uptake of the labeled compound in diseased livers.[154]

Isotopes are also used for studying disorders of the digestive system caused by pancreatitis or biliary obstruction.[155–157]

Gastrointestinal bleeding can be evaluated[158] by labeling the circulating erythrocytes with ^{51}Cr. Erythrocytes labeled with ^{51}Cr have also been used in studies on anemias caused by acute gastroenteritis in the ruminant[159] and in studies on blood compatibility in swine.[160]

Fish. The digestibility of plankton, the acceptance of various-size particles by shellfish, and the function of the blood in transporting various chemicals have been studied with greater ease with isotopes. Tracers have also provided much valuable information on the dispersal of fertilizers in natural waters.[195] Phosphorus-32 can be added to waters and identified in quantities below limits detectable by ordinary methods. Field studies with ^{32}P have confirmed the suspected recycling of organic phosphorus through aquatic food organisms.

INSECT PESTS

Ecology. If known numbers of labeled insects are released and both labeled and normal

specimens are recovered, the rate of dispersal of an insect species, the maximum distance covered in a unit of time, the relative abundance of the insect, and its longevity under field conditions can be determined,[161] and daytime and nighttime resting places can be located.[162] From labeled plants the host preference and nectar requirements of certain nectar-feeding insects can be determined,[163] and from labeled food of social insects,[164–166] the food exchange among colony members can be studied. Mating behavior of insects has been studied by allowing radioactive males to mate with normal wild females; the radioactivity can be measured in the female, and single or multiple matings can be determined.[166] Studies have also been made on movement of olive flies,[167] dispersal of houseflies,[168] migration of mosquitoes,[169] and movement of wireworms in the soil.[170]

Physiology. Radioisotopes have contributed to a better understanding of the physiology and biochemistry of nutrition,[171] circulation,[172] respiration,[173] locomotion, excretion,[174] secretion,[175] reproduction,[176] and nervous-system function. Tracers have been used in studies[177] on the permeability of ^{14}C-labeled glucose and ^{24}Na and ^{42}K through the continuous cellular and fibrous membrane that sheathes the insect nervous system, the biosynthesis of trehalose from ^{14}C-labeled D-glucose in the fat body of the locust,[178] the amino acid sequence in silk fibers produced by silkworms fed on mulberry leaves dipped in solutions of ^{14}C- or ^{35}S-labeled amino acids,[179] and the intermediary metabolism of inorganic ^{35}S into organic ^{35}S in insect tissues.[180]

Control. Radioisotopes have provided an excellent tool for research workers to study the action of insecticides on insects,[181] the detoxification of insecticides by insects and plants,[182] and the persistence of insecticide residues and metabolites in plants and plant products[183] and in animals.[184] An example is the work on the metabolism of ^{32}P-labeled Thiment (phorate) by plants, insects, and animals.[185]

Eradication. Eradication of insects by the sterile-male technique uses the normal mating instinct to eradicate a species without molesting any other species in the environment. The technique is more economical than any other.[186] Radiation has provided an excellent tool for inducing sterility or lethal dominants in many insect species, such as screwworm,[187] housefly,[188] several species of fruit flies, locusts, mosquitoes, tsetse fly, and beetles.[189]

The screwworm was eradicated from the southeastern United States by releasing 50 million sterilized screwworm flies a week, until no egg masses could be found on wounded animals and no normal flies could be collected in traps. The project cost approximately $5 million, but it will save livestock producers approximately $20 million a year.[190]

Reactors and Large Radiation Sources

PLANT BREEDING

The use of neutron irradiation in plant breeding has been limited because of the scarcity of reactors and accelerators, but, as the number has increased, neutron irradiation of plant seeds has also increased. Although less is known about the biological effects of neutron than of X and gamma irradiation, available information about neutron effects indicates that worthwhile results can be expected from more extensive exploitation.[191]

Being more densely ionizing, neutrons are, in general, many times more effective in causing chromosomal disturbances than the sparsely ionizing X and gamma radiations.[192] Their genetic effect is much less dependent on physiological conditions of the cell, oxygen pressure, moisture content, and temperature,[111] and they affect directly the hereditary substance in the cell nucleus with minimum physiological damage.[194] Neutrons have recently been shown to affect specific genes in barley differently from X or gamma rays, and specific desired mutations have been induced for the first time.[121,193,194]

FOOD IRRADIATION

Wholesomeness. Extensive tests have shown no untoward effects in animals or man from eating irradiated foods. In 1963 in the United States, radiation-sterilized bacon and wheat products irradiated for control of insects during storage were cleared for human consumption. Canada had previously permitted the use of ionizing radiation for inhibition of sprouting in potatoes, an application that has also been used on a semiindustrial scale in the Union of Soviet Socialist Republics (USSR). Applications

are being made to public health authorities for clearance on other irradiated products in both North America and Europe.

Disinfestation. Investigations in the United Kingdom and in the United States demonstrated that a radiation dose of 16,000 rads kills all insect species commonly occurring in stored grain with no impairment of the milling and baking properties or decrease in nutritive value.[196] The radiation treatment is practically instantaneous, and disinfestation could be carried out during intake or unloading of grain at a terminal, in contrast to fumigation, which is carried out in static storage.

The International Atomic Energy Agency has outlined a program for a pilot plant for grain irradiation and is investigating the location of a proposed plant. The program includes comparison of isotopes and electron radiation at commercial handling rates for testing efficiency, reliability, economic parameters, and integration problems. A bulk grain irradiator is being constructed in the United States, and in the USSR grain irradiation on a semiindustrial scale has been tested with encouraging results.

Radiation Sanitation. Frozen meats, egg products, and such feedstuffs as meat, fish, bone, and blood meals are among the most likely sources of pathogenic microbes. Radiation offers a promising alternative to destroying the microbes by heat, which is costly and changes the appearance and texture of the products. In London gamma irradiation of imported frozen horsemeat would cost about $14 per ton in a plant treating 13,000 tons/year at 0.65 Mrad (Ref. 197). The cost for radiation control of frozen egg would be similar to that for meat.

Radiation for the elimination of pathogenic microbes in such dried materials as meat or fish meal is under investigation in several countries, and interesting studies are being made on radiation inactivation of foot-and-mouth virus in both moist and dry products.[198]

Radiopasteurization. Early hopes that ionizing radiation could be used to sterilize fresh foods without changing taste, color, or texture have not been fulfilled, but lower dose irradiations have demonstrated the potentials of radiopasteurization as a complement to refrigeration and curing. The keeping quality under refrigeration of haddock, sole fillets, clams, and crab meat is increased manyfold by a few hundred kilorads irradiation.[199] Results with shrimp and oysters are promising. In the United Kingdom this advantage could be achieved at only 2% increase in the retail price.[200] The feasibility of radiation pasteurization of marine products will be further tested on an industrial scale at a 300,000-curie ^{60}Co facility in the United States.

A major problem in any fish-preservation process that does not lead to a sterile product is the potential hazard caused by *Clostridium botulinum.* This dangerous spore-forming organism is exceptionally radioresistant and will survive the doses visualized for radiation preservation of marine products. The factors that influence the radiosensitivity and toxin production of this organism have been studied,[201,202] and it has been concluded that, although *Cl. botulinum* undoubtedly constitutes a potential hazard, this hazard is no greater than with some other accepted commercial products and could be adequately controlled by keeping the irradiated prepacked fish at temperatures near freezing and by indicating a maximum storage life for various products.[200]

Interesting observations have accumulated in a number of countries on the potential usefulness of radiation treatment of fruits. There is considerable agreement on the effectiveness of radiopasteurization of strawberries. The radiopasteurization of sweet cherries, figs, and oranges shows some promise. Results are most promising so far with treatments of a few hundred kilorads.

Large-scale storage and shipping studies of irradiated fruits and vegetables have been initiated by the U. S. Department of Agriculture.

ACTIVATION ANALYSIS

Activation analysis is a highly sensitive and versatile method for determining many elements.[203] Because of the importance of trace elements in agriculture, there is much research on its applicability and versatility.[204–209] A complete radioactivation analytical chart for common trace elements in biological materials has been compiled.[210,211]

Recent improvements in the accuracy and versatility of activation analysis have greatly increased its application in agricultural research. For example, minute quantities of

manganese in flooded rice-paddy soils have been correlated with the productivity of such soils.[212]

Activation analysis has been used to study minerals in biological material[207,213] and to determine trace elements in insects and their origin since the trace-element composition of an insect varies with its habitat.[214]

Neutron-activation analysis is not generally applicable, but other charged particles (alpha, deuteron, and proton) have been used to determine aluminum, beryllium, sodium, and phosphorus. The use of these charged particles is, however, limited by their comparatively small cross-section and the considerable dissipation of heat from the target during analysis.

DESALINATION OF WATER FOR IRRIGATION

There has been considerable interest in the use of heat generated in reactors for desalination of water for agricultural purposes. Although the potential of desalination for providing drinking water is very attractive, the cost of reactor-desalinated water makes its general use for irrigation uneconomical at present. However, for off-season production of high-priced vegetable and fruit crops, this may be feasible in the future.

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Effect of Liming on Availability of Phosphate in "Cerrado" Soils of Brazil

By E. Malavolta, O. J. Crocomo, and R. G. Andrade*

Paper 850 (abstracted)

The effect of limestone on the availability of native phosphorus was studied by the isotopic-dilution method. The available phosphorus in the soil, A, was determined from the equation $A = B(1-y)/y$, where B is the amount of applied phosphorus and y is the fraction of plant phosphorus derived from source B. Numerical values for substitution in the equation were found from rice seedlings to which soil samples and a solution of $KH_2{}^{32}PO_4$ had been supplied as phosphorus source.

A literature survey of agricultural interest and experimental details are given.

New Development in Accelerated Isotopic Labeling of Organic Compounds

By N. A. Ghanem†

Paper 458 (abstracted)

Optimum conditions were studied for tritium labeling of organic compounds by a method based on the use of microwave energy to produce gaseous discharges in radioactive gases. Ions, free radicals, excited states, and other energetic entities produced in the discharge exchange with corresponding atoms in the organic target. The microwave electrodeless discharge allows no sparking on the material, which is at a measurable distance from the discharge zone. Compounds labeled and their specific activities (millicuries per gram) were

L-cocaine · HCl	19	Xylocaine · HCl	1.9
Procaine · HCl	4.8	Astracaine · HCl	64
Tetracaine · HCl	9.6	Carbocaine (base)	22

Experimental details and equipment diagrams are given in the original paper.

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Section

VIII

Isotopes and Radiation Technology

Miscellaneous Items of Interest

The ^{32}P Sources Aboard Gemini III Spacecraft

Sixteen ^{32}P sources were aboard the March 23 Gemini III spacecraft with Major Virgil I. Grissom and Lt. Comdr. John W. Young in a biological experiment to determine the effect of weightlessness and radiation on human white blood cells in culture. The experiment is described in a recent annual report[1] and is briefly reviewed here along with additional details of source preparation and radiological testing. About 200 ^{32}P sources—for preliminary, back-up, ground, and spacecraft experiments—were prepared at Oak Ridge National Laboratory's (ORNL) Isotopes Development Center for the ORNL Biology Division. The Oak Ridge Y-12 Plant designed and fabricated the experimental device, which contained various amounts of activity (^{32}P: 1.71-Mev beta; $T_{1/2}$, 14.3 days) to provide different radiation exposures to blood cells. The device is shown in Fig. VIII-1.

The astronaut exposed the blood samples by depressing the handle of the device after the spacecraft had gone into orbit and the weightless state had been attained. After an appropriate irradiation time, he retracted the handle to end exposure. A duplicate ground-control device was actuated at the same time at the launch site. Radiation effects on blood cells on the ground and in space were being compared at the time of this writing.

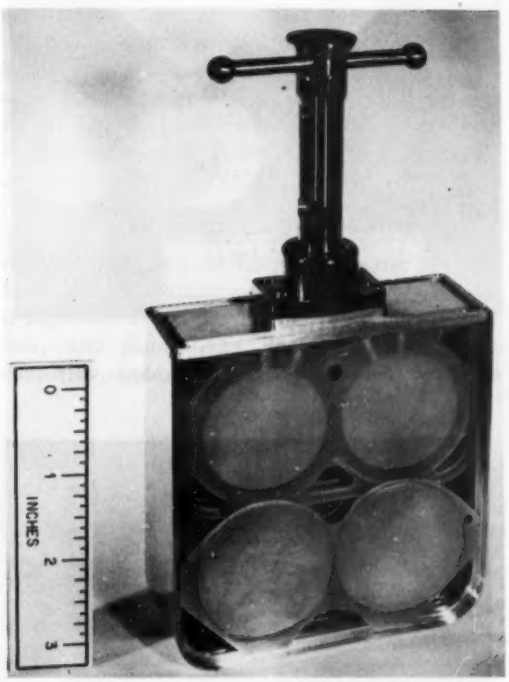

Fig. VIII-1 Assembled Gemini III blood-irradiation device. Side is removed to show the source plates and the blood-sample chambers (in the "up" or non-irradiating position). The sources are shown at bottom, and the blood sample chambers, at top just below the handle. The handle is pressed down to actuate the device.

Source Preparation

The source plates (Fig. VIII-2) were 4-cm disks of 0.003-in. platinum. Platinum was chosen because it is an excellent beta-particle reflector. The increase in bremsstrahlung radiation resulting from the high atomic number of platinum is offset by the fact that the increase in beta flux due to reflection allows the use of less ^{32}P. The ^{32}P used was ORNL stock H_3PO_4 in HCl solution with a radiochemical purity of greater than 99%, exclusive of ^{33}P. Measured volumes of solution were micropipetted onto paper disks (previously cemented to the platinum source plates) in a hot cell (Fig. VIII-3). The ^{32}P concentration was adjusted so that a constant volume of approximately 250 μl was saturated into

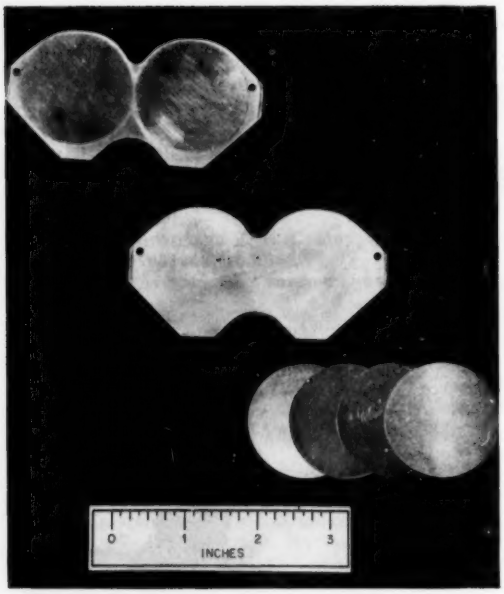

Fig. VIII-2 Source-plate assembly and component parts.

each disk. After about 4 hr of drying, the disks were covered with about 0.5 ml of epoxy resin. Three of the source-plate assemblies had ^{32}P on both sides; the end plates had ^{32}P on one side only. The end source-plate assembly nearest the control blood-sample holder had nonactive disks on the "back" side to help shield the samples from the ^{32}P sources.[2]

Source Testing

The device underwent a series of physical, radiological, and biological tests. Physical tests simulated temperature and environmental conditions, and biological tests concerned the suitability of blood cultures. Radiological tests included measurements of external radiation levels and ^{32}P-source homogeneity.

A complete but unwelded device containing ^{32}P source plates with calculated activities totaling 25 mc (i.e., 16 sources comprising 4 plates each with activities of 2.5, 1.875, 1.25, and 0.625 mc, respectively) was tested for external radiation levels. The radiation field

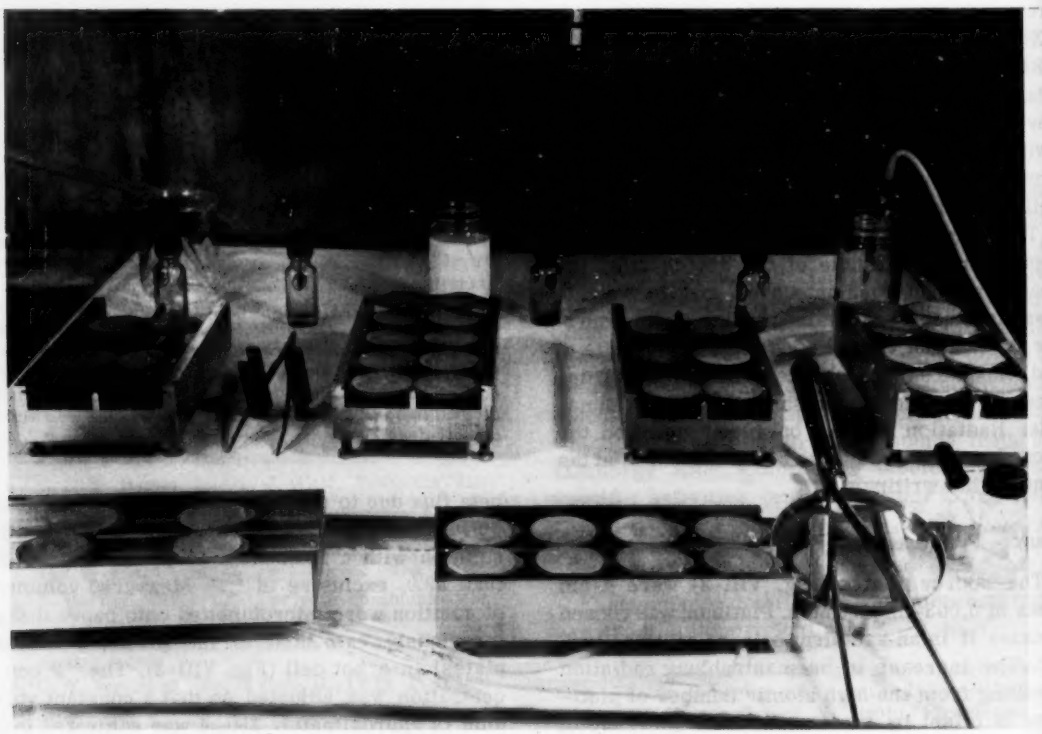

Fig. VIII-3 View of hot cell and source assemblies.

around the device consisted of beta-induced bremsstrahlung as well as beta particles.

Measurements were made with a Thyac G-M survey meter (Victoreen Instrument Co.), which was calibrated against an NBS-calibrated Victoreen electrometer and a 0.25-r chamber. Readings were made at a number of points in each of three planes around the device at a distance of 10 cm, with the beta shield of the Thyac probe in place.

The resulting bremsstrahlung profiles showed this radiation to be less than or equal to 7 mr/hr at 10 cm in any direction and much less at most points.

A test for applicability of the inverse-square relation was also made, and it was found that this relation held for distances greater than 10 cm and could thus be used in calculating the radiation field in the Gemini III vehicle, once the location of the device in the spacecraft was known.

Tests using the Thyac meter with its beta shield removed showed that the beta-particle field around the device was a maximum of three times the bremsstrahlung level on the "hot" side of the device. Since the bremsstrahlung field would be a maximum of about 0.6 mr/hr at 1 ft from the device, it was concluded that there was no radiation hazard to the astronauts or the spacecraft.

The homogeneity of the ^{32}P sources was tested by making autoradiographs of the surfaces of the source-plate assemblies. According to ORNL workers these autoradiographs were valuable indicators of production quality and helped in the refinement of techniques. These tests indicated that the sources were relatively homogeneous and, except for small local defects, did not vary in intensity by more than 20%. An example of such a source is shown in Fig. VIII-4. Fricke dosimetry, spectral analysis, and fluoroglass dosimeter calibration were also made.

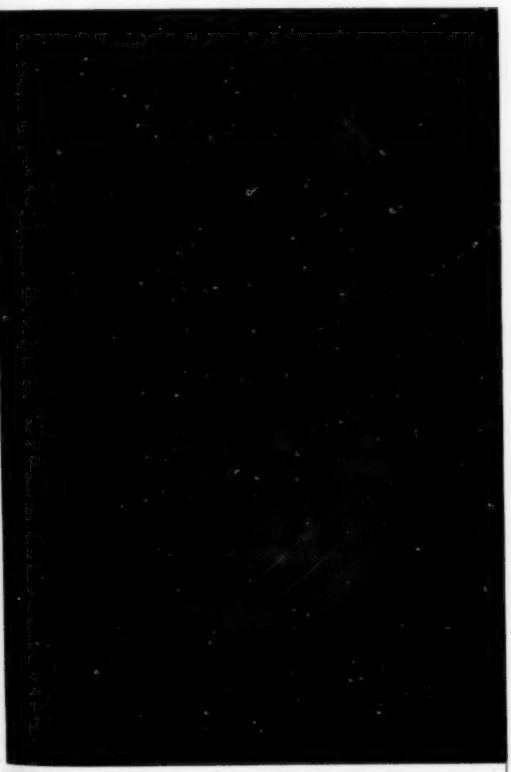

Fig. VIII-4 Autoradiograph of source assembly. Exposure time, 60 sec; activity of ^{32}P, 2.5 mc; 55-speed Polaroid film. The autoradiograph was made after an application of epoxy. Note the homogeneity of the source and the slightly high activity at the edges.

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AEC Activities

AEC Publishes Criteria for Approval of Use of Radioactive Materials in Consumer Goods

The AEC is publishing criteria that it will use in considering applications to use radioactive materials in products intended for distribution to the general public. These applications are evaluated on a case-by-case basis.

A number of consumer products that contain small quantities of radioactive materials already have been approved by the Commission. These include long-standing uses of source material (uranium and thorium), most of which antedate the atomic energy program, such as the use of uranium to color glass and glazes for

decorative purposes; the use of thorium in various alloys and products such as gas mantles, tungsten wire, welding rods, and photographic lenses; and the use of uranium and thorium in photographic film and prints.

The AEC also has approved the use of tritium as a substitute for radium on watch and clock dials and hands, in automobile-lock illuminators, and in precision balances. Uranium is approved as shielding in shipping containers and in fire-detection units.

In approving the use of radioisotopes and of uranium and thorium in consumer products, the Commission establishes limits on quantities or concentrations of radioactive materials and, if appropriate, on radiation emitted. In some cases other specifications important to health and safety, such as quality control and testing, also are delineated.

Information on the total quantities of radioactive materials being used in such products and the number of items being distributed will be obtained through record-keeping and reporting requirements.

A copy of the AEC criteria for approval of consumer products containing radioactive materials follows:

ATOMIC ENERGY COMMISSION

USE OF BYPRODUCT MATERIAL AND SOURCE MATERIAL IN PRODUCTS INTENDED FOR USE BY THE GENERAL PUBLIC (CONSUMER PRODUCTS)

Criteria for the Approval of Products Intended for Use by the General Public Containing Byproduct Material and Source Material

This notice sets forth the essential terms of the Commission's policy with respect to approval of the use of byproduct material and source material in products intended for use by the general public (consumer products) without the imposition of regulatory controls on the consumer-user. This is accomplished by the exemption, on a case-by-case basis, of the possession and use of the approved items from the licensing requirements for byproduct and source material of the Atomic Energy Act of 1954, as amended, and of the Commission's regulations "Licensing of Byproduct Material," 10 CFR Part 30 and "Licensing of Source Material," 10 CFR Part 40.

1. At the present time it appears unlikely that the total contribution to the exposure of the general public to radiation from the use of radioactivity in consumer products will exceed small fractions of limits recommended for exposure to radiation from all sources. Information as to total quantities of radioactive materials being used in such products and the number of items being distributed will be obtained through record-keeping and reporting requirements applicable to the manufacture and distribution of such products. If radioactive materials are used in sufficient quantities in products reaching the public so as to raise any question of population exposure becoming a significant fraction of the permissible dose to the gonads, the Commission will, at that time, reconsider its policy on the use of radioactive materials in consumer products.

2. Approval of a proposed consumer product will depend upon both associated exposures of persons to radiation and the apparent usefulness of the product. In general, risks of exposure to radiation will be considered to be acceptable if it is shown that in handling, use and disposal of the product it is unlikely that individuals in the population will receive more than a small fraction, less than a few hundredths, of individual dose limits recommended by such groups as the International Commission on Radiological Protection (ICRP), the National Council on Radiation Protection and Measurements (NCRP), and the Federal Radiation Council (FRC), and that the probability of individual doses approaching any of the specified limits is negligibly small. Otherwise, a decision will be more difficult and will require a careful weighing of all factors, including benefits that will accrue or be denied to the public as a result of the Commission's action. Factors that may be pertinent are listed in paragraphs 9 and 10 below.

3. It is considered that as a general rule products proposed for distribution will be useful to some degree. Normally the Commission will not attempt an extensive evaluation of the degree of benefit or usefulness of a product to the public. However, in cases where tangible benefits to the public are questionable and approval of such a product may result in widespread use of radioactive material, such as in common household items, the degree of usefulness and benefit that accrues to the public may be a deciding factor. In particular, the Commission considers that the use of radioactive material in toys, novelties, and adornments may be of marginal benefit.

4. Applications for approval of "off-the-shelf" items that are subject to mishandling especially by children will be approved only if they are found to combine an unusual degree of utility and safety.

5. The Commission has approved certain long-standing uses of source material, most of which antedate the atomic energy program. These include:

(1) Use of uranium to color glass and glazes for certain decorative purposes;
(2) Thorium in various alloys and products (gas mantles, tungsten wire, welding rods, optical lenses, etc.) to impart desirable physical properties; and
(3) Uranium and thorium in photographic film and prints.

6. The Commission has also approved the use of tritium as a substitute luminous material for the long-standing use of radium for this purpose on watch and clock dials and hands.

7. The Commission has approved additional uses of byproduct and source material in consumer products. These include the following:

(1) Tritium in automobile lock illuminators;
(2) Tritium in balances of precision;
(3) Uranium as shielding in shipping containers; and
(4) Uranium in fire detection units.

8. In approving uses of byproduct and source materials in consumer products, the Commission establishes limits on quantities or concentrations of radioactive materials and, if appropriate, on radiation emitted. In some cases other limitations, such as quality control and testing, considered important to health and safety are also specified.

PRINCIPAL CONSIDERATIONS WITH RESPECT TO EVALUATION OF PRODUCTS

9. In evaluating proposals for the use of radioactive materials in consumer products the principal considerations are:

(a) The potential external and internal exposure of individuals in the population to radiation from the handling, use and disposal of individual products;
(b) The potential total accumulative radiation dose to individuals in the population who may be exposed to radiation from a number of products;
(c) The long-term potential external and internal exposure of the general population from the uncontrolled disposal and dispersal into the environment of radioactive materials from products authorized by the Commission; and
(d) The benefit that will accrue to or be denied the public because of the utility of the product by approval or disapproval of a specific product.

10. The general criteria for approval of individual products are set forth in paragraph 2, above. Detailed evaluation of potential exposures would take into consideration the following factors, together with other considerations which may appear pertinent in the particular case:

(a) The external radiation levels from the product.
(b) The proximity of the product to human tissue during use.
(c) The area of tissue exposed. A dose to the skin of the whole body would be considered more significant than a similar dose to a small portion of the skin of the body.
(d) Radiotoxicity of the radionuclides. The less toxic materials with a high permissible body burden, high concentration limit in air and water, would be considered more favorably than materials with a high radiotoxicity.
(e) The quantity of radioactive material per individual product. The smaller the quantity the more favorably would the product be considered.
(f) Form of material. Materials with a low solubility in body fluids will be considered more favorably than those with a high solubility.
(g) Containment of the material. Products which contain the material under very severe environmental conditions will be considered more favorably than those that will not contain the material under such conditions.
(h) Degree of access to product during normal handling and use. Products which are inaccessible to children and other persons during use will be considered more favorably than those that are accessible.

(Section 161, 68 Stat. 948; 42 U.S.C. 2201. Administrative Procedure Act, Sec. 3, 60 Stat. 238; 5 U.S.C. 1002.)

Dated at Washington, D. C., this 8th day of March, 1965.

FOR THE ATOMIC ENERGY COMMISSION

/s/ W. B. McCool

W. B. McCool
Secretary

Formal Procedures Adopted for Withdrawal from Routine Production, Sale of Radioisotopes

The AEC has adopted formal procedures for withdrawal from routine production and distribution of radioisotopes that are reasonably available from commercial producers.

The formal procedures by which AEC may withdraw voluntarily, or by which industry might take the initiative to request such withdrawal, were published in the *Federal Register*, Mar. 9, 1965. The procedures became effective upon publication. When the procedures were first published in the *Federal Register*, Sept. 16, 1964, the Commission allowed a 60-day period for public comment. Twenty comments were received from individuals representing 18 organizations.

The procedures and guidelines may be obtained from the Isotopes Sales Department, Isotopes Development Center, Oak Ridge National Laboratory, P. O. Box X, Oak Ridge, Tenn. 37831.

^{147}Pm Illuminators Exempted from Licensing

The AEC recently exempted ^{147}Pm from licensing when the radioisotope is used in automobile-lock illuminators at levels of no more than 2 mc for a single unit. Tritium previously was approved on automobile locks.

Promethium-147 may now be used under a simplified licensing system on aircraft safety

devices such as emergency-exit signs, aircraft switch knobs or plungers, and control locators. The radioisotope may be used on aircraft safety devices under a general license, which is issued without the filing of an application with the Commission. A similar general license now is in effect for the use of tritium on these aircraft safety devices. The maximum amount of ^{147}Pm in a generally licensed aircraft safety device will be 100 mc.

The Commission's action follows receipt of a petition from Minnesota Mining & Manufacturing Co., St. Paul, for an amendment to AEC regulations to permit the use of ^{147}Pm on these items. The AEC requested public comment on the proposed exemptfons and general license in May 1964. These comments were considered in approving the final form of the amendment to AEC regulations, 10 CFR 30, "Licensing of Byproduct Material." The amendment was effective 30 days after publication in the *Federal Register*, Mar. 13, 1965.

Joint Proposal for Hanford Plant Selected

The AEC has selected a joint proposal by Martin Marietta Co. and the United States Rubber Company as the basis for negotiation of a contract for construction and operation of a commercial facility to recover fission products at the Hanford plant near Richland, Wash., and for operating its chemical processing complex.

The new firm formed by the two companies will be responsible for financing construction of the Fission Products Conversion and Encapsulation (FPCE) plant that will operate on a completely commercial basis. The plant will recover usable radioisotopes from partially purified and relatively concentrated solutions received from the Hanford Waste Management Facility.

A conceptual design of a plant capable of processing the Hanford fission products was described in *Isotopes and Radiation Technology*, 2(2): 99-123 (Winter 1964–1965).

General

Russian ^{144}Ce Device Operates A Year

Russia recently announced that Beta-1, a ^{144}Ce-powered automatic weather station near Moscow, has completed a year's operation without maintenance. Scores of these stations will be installed in remote regions, according to the State Committee on Utilization of Atomic Energy. The Beta-1 was developed by the All-Union Scientific Research Institute of Radiation Techniques. The device uses semiconductors to convert heat generated by ^{144}Ce decay into direct current. The temperature, prospective life, or power output of the device were not specified.

New Radiation Device Will Probe Depths

A new radiation device for probing the ocean bottoms may replace time-consuming sampling methods now used in oceanographic research.

The device, a sediment-density meter, is being developed under a U. S. Atomic Energy Commission (AEC) contract by the Lane-Wells Co., Houston, Tex. A unit was tested recently by the Navy in the Gulf of Mexico. It uses scattered gamma rays from a 100-mc ^{137}Cs source.

When the pointed device, 26 ft long and about 4 in. in diameter, is lowered by cable to the bottom of a body of water, its weight drives it into the layered bottom sediments. The traveling detector assembly is triggered to move upward when the meter has penetrated the sediment layer. The assembly then automatically scans the sediments from inside the probe housing. At each stop, measurements are recorded on special cold-resistant film. Since each scan takes only 4 min and the battery power is adequate for 8 hr, investigators can take many measurements before bringing the instrument to the surface. Preliminary tests indicate that the radiation method is more accurate than core sampling since cores expand when brought to the surface.

Ethicon Contracts for New ^{60}Co Unit; UKAEA Licensee To Build Hamburg Plant

A $270,000 irradiation unit will be built for Ethicon G. m. b. H. of Hamburg, West Germany, by H. S. Marsh, Ltd., a licensee of the United Kingdom Atomic Energy Authority (UKAEA), it was announced recently.

This plant will be the sixth commercial ^{60}Co irradiation plant for various Johnson & Johnson subsidiaries. It will be designed for a 750,000-curie ^{60}Co source but initially will be loaded

with about 40,000 curies for sterilizing sutures and other products. The Radiochemical Centre of the UKAEA will supply the radioisotope. The Hamburg contract is the third order for Marsh for such plants, the first being for Johnson's Ethical Plastics Ltd. of Slough, England (like Ethicon, a Johnson & Johnson subsidiary), and the second for Gillette Industries, a subsidiary of Gillette Surgical of Reading, England. Ethicon's facility at Edinburgh, Scotland, was built by Nuclear Chemical Plant Co. Ltd. Ethicon's other ^{60}Co facilities, all built by Atomic Energy of Canada Ltd., are in Somerville, N. J., San Angelo, Tex., and Peterborough, Ontario.

Facilities for Ethicon's previous electron-beam and current ^{60}Co gamma-irradiation plants will be compared in *Isotopes and Radiation Technology*, 2(4): (Summer 1965).

Johnson & Johnson is now the world's largest commercial user of ^{60}Co.

Chief, Aide Chosen in Seibersdorf Project

A project leader and a technical aide recently were appointed for the international food-irradiation program at Seibersdorf, near Vienna.

The project leader, Dr. Karl Kaindl, is the director of the Institute of Agriculture and Biology of the Austrian Atomic Energy Study Company. Dr. Frederick D. Howard, University of California, who is now with the joint FAO/IAEA Division of Atomic Energy in Agriculture, will act as the technical adviser to the project.

These two appointments were announced at the conclusion of the first meeting of a project committee which was attended by representatives from Belgium, France, Federal Republic of Germany, Spain, Switzerland, United States, Austrian Atomic Energy Study Company, IAEA, the Organization for Economic Cooperation and Development (OECD), European Nuclear Energy Agency (ENEA), and by observers from Denmark and Sweden.

The agreement to set up the Seibersdorf food-irradiation program was signed in Vienna on Sept. 16, 1964, by the Austrian Atomic Energy Study Company, ENEA, and the IAEA. Under the agreement, which covers a period of 6 years, fruits and fruit juices are to serve as "model substances" for a research program to determine factors affecting the efficiency of food preservation by irradiation, and the effects (if any) of irradiation on taste and "wholesomeness."

Availability of Isotopes and Services

AEC Withdraws from Routine Production, Sale of Six Additional Radioisotopes

On Apr. 18, 1965, AEC withdrew from the routine production and distribution of six additional radioisotopes—^{45}Ca, ^{59}Fe, ^{125}Sb, ^{75}Se, ^{113}Sn, and ^{65}Zn.

The new action brought to a total of 14 the radioisotopes that AEC has discontinued producing routinely [*Isotopes and Radiation Technology*, 2(1): 93 (Fall 1964)]. As with past withdrawals, however, the Commission continues to supply these radioisotopes when the purchaser certifies in writing that he requires material of a quality that is not commercially available. The six radioisotopes, which are used principally in research and in medical diagnosis, have been produced and distributed by ORNL.

In 1964 AEC withdrew from production of six radioisotopes: ^{141}Ce, ^{58}Co, ^{51}Cr, ^{134}Cs, ^{55}Fe, and ^{85}Sr. In 1963 AEC announced that two others, ^{125}I and ^{131}I, would be produced only when special fission-product material of high specific activity was required. The AEC also continues the policy it established in 1961 of supplying ^{60}Co only when commercial producers cannot meet requirements under conditions of time, quality of product, or reasonable price.

A formal procedure (reported in this issue with "AEC Activities") was adopted this year for AEC withdrawal from routine production.

AEC Announces Changes in Prices of 60 Radioisotopes, Irradiation Services

The AEC increased prices of 52 radioisotopes effective on May 1, 1965, and will reduce prices of 8 others.

The price increases were necessary to recover full costs of radioisotope production and distribution. Some of the more commonly used radioisotopes for which prices were increased are ^{198}Au, ^{47}Ca, ^{131}I, ^{85}Kr, ^{32}P, and ^{89}Sr. Among those for which prices were decreased is ^{14}C, a radioisotope widely used in biomedical research.

The prices for cyclotron irradiation services were increased from $90 per hour to $140 per hour. Commercial processors and distributors

of cyclotron-produced isotopes use this service because no suitable commercially owned machines are available. These companies use the 86-in. cyclotron at ORNL for irradiation of materials that the companies then process for radioisotopes. Modest increases also were made in reactor irradiation service charges.

The AEC gave advance notice of these price changes in keeping with a procedure published in the *Federal Register*, Mar. 9, 1965.

Copies of the revised price schedules may be obtained from the Isotopes Sales Department, Isotopes Development Center, Oak Ridge National Laboratory, P. O. Box X, Oak Ridge, Tenn. 37831, or from Hot Laboratory Division, Brookhaven National Laboratory, Upton, Long Island, N. Y. 11973.

ORNL To Enrich ^{85}Kr by Thermal Diffusion from 5 to 45 Percent for Research Purposes

Oak Ridge National Laboratory is studying the enrichment of ^{85}Kr by the thermal-diffusion process.

The Laboratory expects to produce research amounts of approximately 45% ^{85}Kr from available fission-product krypton gas, which normally contains only about 5% of the isotope. Up to 4000 curies per year will be produced on an experimental basis once the thermal-diffusion columns have reached equilibrium.

The Laboratory shipped 5900 curies of unenriched ^{85}Kr during fiscal year 1964. The isotope, which has a half-life of 10 years, is used in the medical field to trace and estimate the flow of blood through the heart and in industry to detect leaks.

Other established industrial applications include activating phosphors in the manufacture of luminescent signs, estimating temperatures and measuring erosion of turbine blades, determining the wear of bearings, and determining the properties of refractory metals at high temperatures.

In other uses the isotope is introduced into clathrates — compounds such as hydroquinone in which the ^{85}Kr is trapped in the "cage" lattice structure of the organic component — and into substances known as kryptonates. When one of these kryptonated substances reacts chemically with a second substance, the amount of the latter can be estimated by the amount of ^{85}Kr released during the reaction. This technique is suitable for analysis of such substances as sulfur dioxide, ozone, oxygen, fluorine, and hydrogen.

Researchers also believe concentrated, compressed ^{85}Kr may be useful as a heat source.

AECL Reduces Prices for ^{60}Co

Atomic Energy of Canada Ltd., effective Mar. 1, 1965, reduced prices to $0.50 to $0.75 per curie (38 to 57%), depending on quantity, for industrially used low-activity ^{60}Co slugs up to 15 curies/g. Table VIII-1 shows the old and new prices.

At the same time they deleted from the new Price List (PL 15-15 US-EXP) 10 to 54 curies/g ^{60}Co pellets, which are used mainly for industrial radiography and medical teletherapy requirements. Quantity discounts above the new minimum levels of 55 curies/g specific activity remain unchanged for ^{60}Co pellets.

Table VIII-1 FORMER AND NEW AECL PRICES FOR ^{60}Co SLUGS

Specific activity, curies/g	U. S. dollars/curie	Quantity discounts	
		No. of curies	U. S. dollars/curie
May 1, 1963			
0 to 10	1.50	Up to 4999	List price
11 to 15	1.75	5000 to 24,999	List price less 25%
16 to 20	2.00	25,000 to 100,000	List price less 40%
Mar. 1, 1965			
Up to 15	0.75	0 to 4999	0.75
		5000 to 24,999	0.70
		25,000 to 49,999	0.65
		50,000 to 74,999	0.60
		75,000 to 99,999	0.55
		100,000 or over	0.50
		Prices for larger quantities available on request	

Second International Conference on Modern Trends in Activation Analysis

The Second International Conference on Modern Trends in Activation Analysis was held at College Station, Tex., April 19 to 22 and was cosponsored by the International Atomic Energy Agency, Eurisotope Bureau of Euratom, Division of Isotopes Development of USAEC, and the Activation Analysis Research Laboratory of Texas A & M University. About 450 scientists from many countries attended the conference and approximately 70 papers were presented. Subjects of the various sessions included nuclear reactions, radiochemistry, chromatography, neutron generators, instrumentation, computer methods, and biological and industrial applications. The conference also included an exhibit of commercial activation-analysis equipment—accelerators, neutron generators, multichannel analyzers, neutron detectors—as well as USAEC's activation-analysis exhibit.

Complete proceedings of the conference will be published by *Nucleonics*.

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